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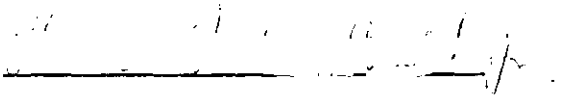

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RADIOTRACER STUDY OF RAPID SAND FILTRATION

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SUMMARY

Rapid sand filtration involves flow through a granular bed of material at rates on the order of two to five gallons per minute per square foot of filter surface. It is a well known commercial process that has been widely used for many years. Although design and operation of rapid sand filters has been refined to a high degree through trial and error, the exact scientific laws governing the process have not been precisely elucidated. Many possible mechanisms by which particles may be removed from flowing water during passage through a porous medium have been suggested; the problem is to determine which of these mechanisms or combination of them produces the observed effect.

One aspect of the process is the relationship of the size of the suspended particle to the efficiency of filtration. Another factor involved is the size of the suspended particle relative to the size of the grains comprising the porous bed. The effect on efficiency of filtration of the relationships of dimensions of suspended particles and bed grains was studied experimentally by determining the density of particle deposition at fixed elevations within a filter matrix. This was accomplished by use of vermiculite particles labeled with radioactive cesium-137. Labeled particles in narrow size ranges were suspended in water and passed into a laboratory-scale filter built to simulate the construction of a commercial filter. The radioactive particles were followed with radiation detection equipment. Three differ-

ent sizes of sand and one size of ground anthracite were utilized as the filter matrix.

The observed pattern of deposition was found to depend on both the absolute and relative sizes of the suspended particles and the stationary grains. From the pattern of particle deposition, an efficiency parameter, the filter coefficient, was calculated. In a given filter medium, the efficiency of particle removal was found to vary directly with the diameter of the particle. For a given particle size, removal efficiency was found to vary inversely as the diameter of the grains of the filter bed. The relationship of filter coefficient to particle diameter is not linear over the complete range of sizes studied, but the departure from linearity of the filter coefficient of particles with diameters less than about 20 μ is explained on the basis of entrapment by van der Waals and electrical double layer forces, whereas larger particles are trapped by interstitial sieving.

The functional interdependence of grain and suspended particle dimensions is such that the filter coefficient varies inversely in a linear fashion with the ratio of pore diameter to particle diameter for ratios of about five or less. Experimentally measured parameters were compared with values predicted by theory associated with the sedimentation hypothesis of filter action. According to this hypothesis, only gravitational force is needed to explain removal of particulates; this idea is rejected on the basis of the experimental evidence for the conditions involved.

Dissolved cesium-137 was found to be sorbed reversibly by quartz

sand but is sorbed irreversibly by ground anthracite. A filter containing sand may be washed free of dissolved cesium-137 ion by flowing water, but this is not possible if the filter medium is ground anthracite.

CHAPTER I

INTRODUCTION

General

The separation of solids from liquids by filtration is a natural phenomenon that occurs whenever a particle-carrying liquid encounters a permeable membrane or material. It occurs continuously in the life of plants and animals during the movement of internal fluids, and in the natural environment where sediment-carrying streams pass over porous formations, where raindrops containing dust particles strike the ground and then infiltrate into the soil, and where subterranean flows encounter permeable formations. Filtration by soil is largely responsible for the generally low turbidity of groundwater and man has been copying this process since he discovered that clean water is more palatable than dirty water.

Filters were in use before the beginning of recorded history; they were described by ancient Chinese and Hebrew writers and were pictured in Egyptian tombs.[1,2] Other writers in the first few centuries A. D. described earthen vats with perforated bottoms that supported grass mats, cloth, or a bed of lime.[3] The Middle Ages saw few improvements and when interest was revived by industrial developments in the 18th century most of the earlier technology had been lost and a new start was necessary.[4]

Joseph Amy received one of the first French patents in 1789.[5]

It was for the purification of Paris drinking water by "passing it downward by gravity through sand in false-bottomed household containers." Two years later Peacock received the first British filter patent for a similar process which also included a system of backwashing.[6] This backwashing feature eliminated the need to remove the filter medium for cleaning and led to the construction of many large sand filter beds for municipal water purification. Filters became even more popular after it was discovered that they remove a large fraction of the bacteria present. From these beginnings, the technology of sand filter operation has been developed to a high degree.

In addition to the traditional problems of water purification, today we are faced with the additional problem of an increase of radioactivity over natural background levels in our water. Radioactivity has always existed on the earth and at low levels in natural waters, but only the last 20 years have seen the production, concentration, utilization, and concomitant need for disposal of radioisotopes rise to their present level, and further large increases in production and use will undoubtedly follow.

A basic tenet of the life sciences is that excess radiation is harmful to all life. While it is not possible to avoid contact entirely with radiation, exposure should be kept to a minimum by careful control of concentrated quantities of radioactive materials. Disposal of radioactive wastes should be carried out in such fashion as to insure minimum contamination of the air, water, food, and other portions of the environment of man. There are two practical possibilities: disposal on land or in the ocean. Land disposal is preferred because the site

for accumulation of hazardous concentrations can be selected and controlled.

Much dilute radioactive waste is presently being released to surface streams, and although there is no particular hazard now, this will ultimately lead to contamination of the oceans and all forms of marine life. The alternate to immediate release of a waste is a period of storage sufficiently long to allow radioactive decay to reduce the activity to levels acceptable for release to the environment. In the case of long-lived isotopes, storage may be required for centuries, and one problem this presents is the continuing possibility of containment failure which might result in severe contamination of the local groundwater or nearby streams.

The radionuclide level in streams may possibly increase due to the discharge of reactor cooling water, reactor fuel reprocessing, medical and industrial uses of radionuclides, or from fallout resulting from nuclear explosions above or below ground or from accidental emission from a reactor. Any such increase in the level of radionuclides would place an even greater responsibility on those concerned with public water supplies. Even under normal conditions, operators of water treatment plants have a more urgent responsibility than the operators of other utilities because of the indispensibility of water. We could probably survive without the modern accoutrements of civilization such as electricity, gas, and telephones, but not without a continuing safe water supply. It is therefore important that the water treatment plant operator not only be competent to produce a satisfactory product under normal conditions, but that he also plan ahead for possible emergency situations.

One such emergency would be the sudden appearance of a high level of radioactivity in the raw water supply. This could be brought about by an upstream spill of radioactive material from an industrial plant, from a truck carrying such material running off a bridge, or numerous other possible events. Such occurrences seem rather remote, but the growing distribution and use of radioactive materials makes them increasingly less improbable. It would therefore be advantageous to evaluate the possible consequences of a situation of this sort.

It is not possible to foresee all contingencies, and therefore detailed procedures for all eventualities cannot be developed in advance. It is, however, possible to evaluate the response of various plant components to abnormal conditions and be prepared to make knowledgeable decisions based on information gathered and assimilated under non-emergency conditions. Rapid sand filters are one of the more important plant components to be considered in such a study.

Radioactivity in water will be present in suspended form, dissolved form, or probably both forms. In fallout the radionuclides are generally associated with particulate matter while industrial effluents are more likely to contain dissolved activity. In either instance, the radionuclides entering a stream may dissolve or remain in solution; however, a significant amount will become associated by sorption or ion exchange processes with the mineral sediments, organic debris, and aquatic organisms. The relative distribution of the radionuclides will depend upon the isotope under consideration and the environmental factors.[7] The attachment of radioactive atoms to a particle does not detectably affect the behavior of the particle, as long as the size or chemical

nature of the particle remains unchanged. The presence of radioactive particulate matter will therefore not affect the rapid sand filtration process. Removal will occur in the same fashion, and with the same efficiency as non-radioactive matter.

A dynamic equilibrium is established between dissolved ions in the liquid and those immobilized on the surfaces of the filtration medium, and no further net change occurs as long as the ion concentration in the water remains constant. Barring complete disaster, radioactive ions would be present only in very low concentrations, and even a small change in concentration might be highly significant. Since filtration is such an important means of particle removal, a full understanding of the mechanism of removal involved is of practical as well as theoretical interest. While much is known empirically about the operational aspects of the filtration process, it is evident that the mechanism is still not fully understood; it is the purpose of this work to provide additional experimental evidence and an interpretation leading closer to a more complete understanding. Apart from radioactive decontamination, a thorough understanding of the filtration process is clearly desirable in view of its many applications in sanitary, chemical, and military engineering as well as in agricultural processes. Such a study, involving the attainment of quantitative information on the removal and retention of fine particulate matter, can best be carried out by the use of radioactive tracers.

A porous material may be described as a solid that contains numerous holes or voids distributed within it in an ordered or random fashion. In materials such as sandstone or pumice, the solid is con-

tinuous; a bucket of sand is an example of a discontinuous or unconsolidated solid. The holes may or may not be interconnected, but the subject here involves the passage of fluid through the material, and consideration will be limited to unconsolidated matter with interconnected holes.

More specifically, the porous materials considered in this study were sand and ground anthracite through which water containing suspended particles was passed. Such "beds" of granular matter are characterized by a number of physical and geometrical variables including porosity, packing, grain shape and size, and grain size distribution.

Theories have been advanced which attempt to relate "pore size distribution"[8] or "grain size distribution"[9] of unconsolidated materials to the statistical properties of their microscopic structure. Such theories have not been totally satisfactory and the usual approach to the matter is through the macroscopic laws as established by experiment.

The "porosity" of a porous material is the fraction of the bulk volume of the material occupied by voids. Porosity, P, is, therefore, the ratio

$$P = \frac{\text{Volume of pores}}{\text{Bulk volume}} \tag{1.1}$$

which is a dimensionless quantity. There are two classes of porosity: "absolute" (or total) and "effective" porosity. Absolute porosity is defined by the above expression without regard to pore interconnections, while effective porosity is the fraction of bulk volume composed of interconnected voids. In filtration the voids may be filling up with

deposits of solid matter, and in such cases the term "effective volume" is not a fixed quantity, but refers to the fraction of the void volume remaining interconnected and permeable at the moment of consideration.

Porosity of an unconsolidated material depends on several things including the degree of compaction and the manner of packing. Compaction is the process of volume reduction due to an externally applied pressure or vibration. Pressure has an appreciable effect on the porosity of compressible materials. Sand, however, is a rigid solid and pressure has little effect unless it is high enough to crush the individual grains. In sand filters gravity and the force of the liquid flow are the forces aiding compaction. Filters are normally cleaned by reverse liquid flow at a rate sufficient to raise and suspend the grains in the liquid while they are tumbled and rubbed against each other by the turbulent backwash flow. This effectively loosens any fine trapped matter which is then washed out. When the reverse flow is stopped the sand settles back into position.

The shapes of the openings are frequently idealized as uniform volumes formed by tangent spheres in an ordered packing arrangement. An alternate approach views them as a bundle of capillaries, but neither description accurately represents the situation that really exists. Individual grain shapes may deviate considerably from spheres; they are irregular and may be angular, and the interstices vary in size and configuration. The capillary analogy is useful, but only with the realization that the openings are most likely not circular, the walls converge and diverge irregularly, and there are numerous lateral interconnections. A study[10] of six different packings of uniform spheres revealed poros-

ities ranging from 0.2595 for rhombohedral packing to 0.4764 for cubic packing. Such studies are useful even if the resulting pores are extremely simple when contrasted with those of a natural material. Theoretically, for uniform spheres, porosity should be independent of the size of the spheres, but with typically non-spherical natural materials this is not the case. Experimentally it is found that with sands of uniform grain size the porosity increases as the grain size decreases due to the greater tendency to "bridging" of the smaller particles, and this is generally true with most naturally occurring unconsolidated materials. In cases where the grain size is non-uniform, the porosity depends on the distribution of sizes, but is lower than with unisize grains as the smaller particles can fill the interstices formed by the larger grains.[11]

The "specific surface" of a porous material is defined as the interstitial area of pore surface per unit of bulk volume of porous material. As the surface of a sphere is proportional to the square of its diameter, fine grained materials possess a higher specific surface area than coarse materials, and this is an important parameter in the derivation of fluid conductivity or permeability due to the significant effect the walls have upon hydrodynamical phenomena. As an area divided by a volume, specific surface has the dimensions of reciprocal length.

"Permeability" is that property of a porous material which characterizes the ease with which a fluid can be made to flow through the material by an applied pressure gradient. It is an inverse flow resistance and is frequently referred to as fluid conductivity. Numerical values of permeability are derived from the relationship given by Darcy's

law which is discussed below. The "darcy" is the usual unit of permeability and is defined as follows: for a material of one darcy permeability, a pressure differential of one atmosphere will produce a flow rate of one cubic centimeter per second of a fluid with one centipoise viscosity through a cube having sides one centimeter in length.

$$1 \text{ darcy} = \frac{1 \text{ (cm}^3/\text{sec)} \cdot 1 \text{ (cp)}}{1 \text{ (cm}^2) \cdot 1 \text{ (atm/cm)}} = (\text{cm}^2 \cdot \text{cp} \cdot \text{sec}^{-1}\text{atm}^{-1})$$

For low values, the millidarcy (10^{-3} darcy) is used.

Naturally occurring porous materials have a more or less random distribution of pore sizes and positions, and samples from different sources are likely to exhibit wide variations in distributions. Ranges of values are given in Table 1 to illustrate the characteristics of a few representative porous materials.[11]

In a water supply undesirable components may be present in various conditions of dispersion. They may be dissolved, in the colloidal state, emulsified, or suspended. Virtually all natural waters contain bacteria and dissolved substances. In addition, surface streams commonly carry silt ranging in size down to colloidal dimensions, and downstream from the domestic or industrial activities of man one finds evidence of human and industrial wastes and is likely to find fats, oils, dyes, detergents, and chemicals of an almost endless variety. Swamp waters and other stagnant or slow-moving bodies of water are likely to contain high concentrations of hydrogen sulfide and particles of organic matter released by decaying vegetation; algae will usually be present. Ground waters usually contain less suspended or colloidal matter, as

Table 1. Physical Characteristics of Typical Porous Materials

Substance	Porosity (fraction)	Specific surface (cm^2/cm^3)	Permeability (Darcys)	Reference
Silica powder	0.37 - 0.49	6.8×10^3 - 8.9×10^3	1.3×10^{-2} - 5.1×10^{-2}	12
Loose sand	0.37 - 0.50	1.5×10^2 - 2.2×10^2	20 - 180	12
Soils	0.43 - 0.54	2×10^3 - 4×10^3	29 - 140	13
Sandstone	0.08 - 0.38	1.5×10^4 - 10×10^4	5×10^{-4} - 3.0	14
Limestone	0.04 - 0.10	0.15×10^4 - 1.3×10^4	2×10^{-4} - 4.5×10^{-2}	15
Brick	0.12 - 0.34	3×10^3 - 5×10^4	4.8×10^{-3} - 2.2×10^{-1}	16
Leather	0.56 - 0.59	1.2×10^4 - 1.6×10^4	9.5×10^{-2} - 1.2×10^{-1}	17
Fiberglass	0.88 - 0.93	5.6×10^2 - 7.7×10^2	24 - 51	18

these are removed by the bounding materials during subsurface movement.

When water containing dissolved ions encounters a sand bed, ion exchange or sorption processes may occur, but when equilibrium is attained, continued filtration has no further effect on ion concentration. If particulate matter is to be removed from a water, filtration is the almost universally practiced process.

There are two general types of filtration: the single collision type that occurs when the filter consists of a thin sheet of perforated or porous material and the multiple collision type (deep filter) achieved when the filter consists of an appreciable thickness of a porous, fibrous, or granular matrix. The single layer type of filter has found only limited use in water treatment. It does an effective job in those occasional instances when the matter to be removed is relatively large and uniform in size, as for example, algae. The deep filter is much more frequently applied because it not only removes such specific entities, but a wide variety of others as well. These two types of filter serve different purposes and the mechanisms by which they operate are different. While the many similarities make it difficult to discuss one without the other, the main interest here centers on the deep filter.

Deep filters are most commonly found in water treatment plants where they consist of large concrete boxes containing coarse sand. Water passes through the sand as one stage of the purification process for municipal or industrial use. These filters are spoken of as sand filters, and this terminology is so firmly fixed that it is retained even for water filters containing beds other than sand.

Sand filters are also classified arbitrarily and rather indefi-

nitely according to the velocity at which the water passes through. While there is no exact dividing line, rates on the order of 0.1 gal/-ft^2/min are termed slow, and rates on the order of one gal/ft^2/min or higher are considered rapid. The slow sand filter produces an effluent of low turbidity and high quality, but is obsolescent in most areas of the world due to the large filter areas required to provide sufficient water quantities for the needs of today. Although somewhat less effective than the slow sand filter, the rapid sand filter can furnish effluent of the same quality when handling properly prepared influent.

From the latter part of the 19th century until the 1930's, major efforts in sand filtration were exerted to optimize operational procedures and refine the mechanical appurtenances.[19] As a result, the art of filter operation has been developed to a very high degree. During this time, filter design became standardized and was based mainly on the hydraulic conditions involved. Little, if any, attention was directed toward the quality of the raw material, a situation that must be unique among industrial processes.

Once the practical technology of filter operation had been developed, interest began to arise in the theoretical aspects of sand (and other) filters. Early investigators of theory include Izbash (1933),[20] Patrashev (1935),[21] Baylis (1937),[22] and Iwasaki (1937),[23] although the study of flow through porous media goes back at least to Darcy (1856).[24]

The mechanism of transport and deposition of fines in porous media has been studied mostly by sanitary engineers interested in sand filters and by soil scientists interested in the effects of horizontal layers of differing types of soil on water and material movement, but

other disciplines are also represented. As a result, the terms, symbols, and concepts of different authors vary considerably, and a comparison of conclusions is difficult, particularly where units were not precisely defined. In the following literature review, an attempt is made to achieve a consistent terminology and a set of consistent symbols. By definition, the term "filter" shall mean the porous matrix through which the filtrate passes.

When turbid water is passed through a filter, the turbidity diminishes, and suspended particulates are deposited in the filter. The exact mechanism by which this occurs is still relatively obscure, although many aspects of the associated phenomena have been thoroughly investigated. The following discussion will cover much of this pertinent information and will set forth the merits of the various mechanisms so far suggested. It now seems that no one mechanism can account for all the experimental evidence and, indeed, there probably exists a combination of forces that produce the observed effects.

Theory of Filtration

The flow of water through granular beds was studied by Darcy,[24] whose work resulted in the familiar equation

$$q = KJ = KH/L \qquad (1.2)$$

where q = specific flux or discharge (rate of flow per unit area)

K = permeability of the matrix, now called hydraulic conductivity, usually expressed in darcys

J = hydraulic gradient or head loss (H) per unit filter length (L)

The evaluation of the permeability coefficient is of interest and led to an empirical equation by Hazen[25] which has found some use

$$K = Cd_{10}^2 \tag{1.3}$$

where d_{10} = "effective size" of sand, which is the sieve size (mm) through which 10 percent of the material passes and 90 percent is retained

C = a size coefficient, approximately equal to 100 but varying between 41 and 146 for sand sizes between 0.1 and 3 mm and with a uniformity coefficient less than 5. The uniformity coefficient is the ratio of sizes of the 60 percentile to the 10 percentile of the distribution.

As all the liquid within a given matrix volume does not move at the same rate, Dupuit[26] defined an average velocity as

$$V_e = q/n_e \quad ; \quad n_e = n - n_o \tag{1.4}$$

where n = total porosity of the matrix

n_e = effective porosity

n_o = porosity of non-contributing void volume containing stationary liquid

It has been reported by Irmay[27] that n_o is usually about 0.25n.

The Darcy relationship is strictly true in steady flow only when the permeability K is independent of time, space, direction, pressure, specific flux, hydraulic gradient, filter length, and cross sectional area.[28] To achieve such idealized conditions, the porous matrix must

be homogeneous, isotropic, and physically, chemically, and geometrically stable. It must have interconnected pores saturated with gas-free liquid and there must be no biological activity. The liquid must be isotropic, homogeneous, incompressible, at constant temperature, density, and viscosity, and physically and chemically inactive. The flow must be slow (in the laminar regime) and there must be no large passages along the container walls to produce an edge effect or channeling.

Under these restrictions, permeability depends on the kinetic viscosity ν of the liquid and the texture of the porous matrix which may be expressed by its porosity, structure and grain shape, specific surface (σ_m) or effective diameter (d_m). These factors have been expressed by Carman[29] as

$$K = a \frac{g}{\nu} d_m^2 \frac{n^3}{(1 - n)^2} \tag{1.5}$$

where d_m = the mean diameter in a graded mixture of spheres

a = a numerical coefficient dependent on the matrix grain shape and packing

g = the gravitational constant.

Agreement between theory and experimental results was improved by a slight refinement by Irmay[30] in which n is replaced by $(n-n_o)$ with an appropriate change in the numerical coefficient

$$K = b \frac{g}{\nu} d_m^2 \frac{(n - n_o)^3}{(1 - n)^2} \tag{1.6}$$

For spheres, a was found to have a value of about 1/180 and b about 1/80.

These formulas[28] may be obtained by comparing laminar flow through a porous medium with flow through a circular tube of diameter D. Using the Poiseuille formula,[31] the mean velocity is

$$V = Jg\ D^2/32\ \nu \qquad (1.7)$$

The hydraulic radius R is defined as the liquid volume of the circular tube divided by the area of the corresponding wetted solid surface. In a circular conduit flowing full, $D = 4R$, and substituting

$$V = Jg\ R^2/2\ \nu \qquad (1.8)$$

In a porous medium

$$R = \frac{n}{(1 - n)\ \sigma_m} \quad ; \quad \sigma_m = \frac{6}{d_m} \qquad (1.9)$$

Substituting into the Darcy equation (1.2), the resulting equation for permeability, K, is 1.5 if n_o is neglected or 1.6 if n_o is considered. These formulas correlate well for the laminar flow of clean water through porous media, but a different situation exists in filtration due to the deposition of solids. Many of the physical and physico-chemical properties of a filter are changed, including the porosity, density, hydraulic conductivity, surface properties, and so forth. Constriction of the pores alters the flow velocity and possibly other flow characteristics.

Over the years a number of investigators have extended the usefulness of Darcy's law to conditions other than the idealized case described above. The conditions include non-homogeneous media,[32,33]

non-isotropic media,[34,35,36] geometrically unstable media,[37,38] unsteady flow,[39,40,41] compressible fluids,[14] unsaturated media,[42,43,44,45] and solutions and adsorptive media.[46] Application of theory to various problems has been discussed in detail by Muskat,[14] Polubarinova-Kochina,[41] and Scheidegger.[47]

Numerous empirical formulas have been devised to describe the filtration process. A frequently encountered but not very useful example was developed by Fair,[48] based on data of a committee of the American Society of Civil Engineers.[49] For a terminal head loss of eight feet, an effluent containing 0.2 mg/l of turbidity or less, and a constant flow rate of two gal/ft^2/min

$$L = K' d^{1.87} \left(\frac{60}{T + 10}\right) \tag{1.10}$$

where L = required filter depth (inches)

d = sand grain diameter (mm)

T = water temperature (degrees Fahrenheit)

K' = a coefficient depending on water quality

An equation proposed by Stanley,[50] also for an eight foot terminal head loss, but at a fixed temperature (25°C), is known to apply only to ferric floc

$$P = K'' d^{2.46} Q^{1.66} \tag{1.11}$$

where P = depth of penetration of ferric floc (inches)

d = sand grain diameter (mm)

Q = rate of filtration (gpm/ft^2)

K'' = 6.4 for the units given

Equations such as these do not give any information on the variation with time of either the filtrate quality or the head loss, and since they are empirical, they can be safely applied only under the specific conditions for which they were derived.

There are certain physical aspects of filtration that can be easily demonstrated. As set forth by Iwasaki[23] and later amplified by Stein,[51] these may be stated: 1) filtration is a dynamic process, filter action being dependent on depth of the filter and velocity of flow through the filter; 2) removal of suspended particles at any depth of the filter is proportional to the concentration of particles at that depth; 3) the coefficient of this proportionality first increases linearly then decreases nonlinearly with the amount of clogging; and 4) material removed from suspension clogs the filter pores. These seemingly simple statements have led to considerable experimental and mathematical complexities.

Using the term "quality" to denote the concentration of suspended matter, a complete mathematical model, given a filter of known characteristics and required output, should accurately predict the variation of filtrate quality with influent quality, the variation of filtrate quality with time, and the pattern of pressure loss due to the accumulation of deposited solids in the pores. The closest approach to this capability has been made by Ives[52] in his modified filter coefficient theory. His analysis was made for the following conditions.

1. The suspended material in the influent is discrete, homogeneous, unisize, denser than the fluid, and about two orders of magnitude smaller than the filter pores.

2. The filter is granular, isotropic, homogeneous, and unisize.

3. The fluid is in laminar flow.

Two equations suggested by Iwasaki[23] form the starting point

$$-\frac{\partial C}{\partial L} = \lambda C \tag{1.12}$$

and

$$\frac{\partial \sigma}{\partial t} + \frac{v}{(1 - f_\sigma)} \frac{\partial C}{\partial L} = 0 \tag{1.13}$$

where C = volumetric concentration of material entering a unit volume of filter

L = filter depth

λ = coefficient of proportionality (also called the filter coefficient)

t = filtration time

σ = volume of suspended material retained per unit of filter volume

f_σ = porosity of the deposited material

v = superficial filtration velocity

Equation 1.12 states that removal of particles is a first order reaction; that is, it is proportional to the concentration of particles present. Most other investigators in the field of water filtration have made this same assumption, and a rational basis for it has been provided by investigators in aerosol filtration.[53]

Equation 1.13 expresses continuity by stating that the volume of material deposited in the filter equals the volume removed from suspension. This assumes that the density and porosity of the deposited ma-

terial do not change during the filtration run, and biological and chemical reactions do not cause soluble materials to either accumulate or be released from the deposits.

It has been found that the filter coefficient is not constant but is altered by the accumulation of deposited matter during filtration. Hence Ives suggested the empirical expression for λ

$$\lambda = \lambda_o + c\sigma - \frac{\phi\sigma^2}{(f - \sigma)} \tag{1.14}$$

where λ_o, c, and ϕ are constants, and f is the initial filter porosity. This equation correlates the variation of the filter coefficient with the specific deposit, σ, the volume of deposited material per unit filter volume. Substituting 1.14 into 1.12 yields

$$-\frac{\partial C}{\partial L} = \left(\lambda_o + c\sigma - \frac{\phi\sigma^2}{(f - \sigma)}\right) C \tag{1.15}$$

These equations are not amenable to explicit solution, but they have been solved numerically by means of a computer.

Ives[52] further proposed that

$$\frac{\partial h}{\partial L} = \left(\frac{dh}{dL}\right)_o + \bar{k}\sigma \tag{1.16}$$

where h is the head loss, $(dh/dl)_o$ is the head loss per unit depth at $t = 0$ ($\sigma = 0$), and $\bar{k}\sigma$ is a head loss constant. This equation was based on experimental observations and describes the dependence of the hydraulic gradient on the specific deposit. Equations 1.13, 1.14, and 1.15

describe the changes in concentration and specific deposit with time and position; combined with 1.16 they describe head loss changes with time and position. Curves computed with these equations have correlated well with experimental curves obtained in the laboratory.

Unfortunately, the application of these equations to practical situations requires values for five constants. One of these, f_σ, may be only a characteristic of the material in suspension, but λ_o, C, ϕ, and k are related to the nature of the filter, the manner of its operation, and the suspension being filtered. In order to use these equations, it would be necessary to determine experimentally values for these five constants for each set of circumstances to be considered.

Ives and Sholji[54] have studied the effects of sand grain size (d), porosity, flow rate (v), and water viscosity (μ) on the parameters λ_o, C, and ϕ. From theoretical considerations they deduced that

$$\lambda \propto \begin{Bmatrix} \lambda_o \\ C \\ \phi \end{Bmatrix} = (\text{constant}) \frac{1}{d} \cdot \frac{1}{v} \cdot \frac{1}{\mu^\gamma} \tag{1.17}$$

and they experimentally determined the value of (constant) and of γ for each of the three parameters. They found for their particular conditions

$$\lambda_o \propto \frac{1}{dv\mu^2} \tag{1.18}$$

$$C \propto \frac{1}{dv\mu^{1.2}} \tag{1.19}$$

$$\phi \propto \frac{1}{dv\mu^2} \tag{1.20}$$

Other authors have also attempted to develop theoretical relationships between filter performance and these same parameters, and a comparison of results is instructive. Stein[51] reported that the filter coefficient was not dependent on the velocity of filtration or water viscosity, but he proposed the following equation based on the geometry of the constrictions in filter pores and the deposits around the grains

$$\lambda \propto \frac{1}{d^3} \tag{1.21}$$

Hall[55] developed two equations based on filtration by a straining mechanism and also by analogy with settling

$$\lambda \propto \frac{1}{d^{1.5}} \text{ (straining)} \tag{1.22}$$

$$\lambda \propto \frac{1}{\mu v d} \text{ (settling)} \tag{1.23}$$

Mints and Krishtul[56] derived the following equation partly from dimensional analysis and partly empirically

$$\lambda \propto \frac{1}{v^{0.7}\, d^{1.7}} \tag{1.24}$$

They noted that water viscosity was a variable but did not incorporate it into their work.

An inverse proportionality of filter coefficient and velocity was offered by Maroudas;[57] it was based on a consideration of the constriction of flowpaths in filter pores. No theory was offered on the significance of grain size or water viscosity

$$\lambda \propto \frac{1}{v} \tag{1.25}$$

Hydrodynamic forces and van der Waals forces were considered by V. Mackrle and S. Mackrle[58] who suggested the relationship

$$\lambda \propto \frac{\mu^{\frac{1}{2}}}{vd^2} \tag{1.26}$$

The effect of viscosity involved here is in disagreement with other researchers. It implies that filter performance becomes less effective as the temperature of the water rises, and this is clearly contrary to the findings of others.

By rearranging and reinterpreting some of the empirical relationships available, they can be put into comparable form. The report of Fair[48] yields

$$\lambda \propto \frac{1}{\mu d^{1.67}} \tag{1.27}$$

and Iwasaki's data[23] can be reworked to yield

$$\lambda \propto \frac{1}{vd} \tag{1.28}$$

From the research of Stanley,[50] the following relationship can be deduced

$$\lambda \propto \frac{1}{v^{1.56} d^{2.46}} \tag{1.29}$$

Ling[59] used a constant flow rate in a series of experiments and concluded that, for a given turbidity removal, the required depth increased as the

grain size of the bed increased. The expression is

$$\lambda \propto \frac{1}{d^{1.5}} \tag{1.30}$$

The work reported by Hudson[60] leads to the proportionality of the logarithm of a reciprocal function of the three parameters

$$\lambda \propto \ln \frac{1}{F(v,\ d^3,\ \mu)} \tag{1.31}$$

All of these equations may be reduced to a single form

$$\lambda \propto \frac{1}{v^{\alpha}\ d^{B}\ \mu^{\gamma}} \tag{1.32}$$

and perhaps this indicates a general conformity of results, although the wide diversity of values for α, B, and γ leads others to the opposite conclusion. As the various experimental conditions differed in almost every respect except flow regime (they were all run with laminar flow), there is no obvious reason why the detailed values of these constants should coincide. Indeed, it seems that they are dependent on the many variable characteristics of the suspensions, the filter medium, the experimental apparatus, and perhaps the technique of the investigator.

In addition to efforts to define exactly the relationship of λ to different variables, the geometry of particle deposition has been examined. Three mathematical models, based on different physical concepts, have been proposed and studied by Sakthivadivel.[61] A definition sketch, given in Figure 1, illustrates the model which is based on a visualization

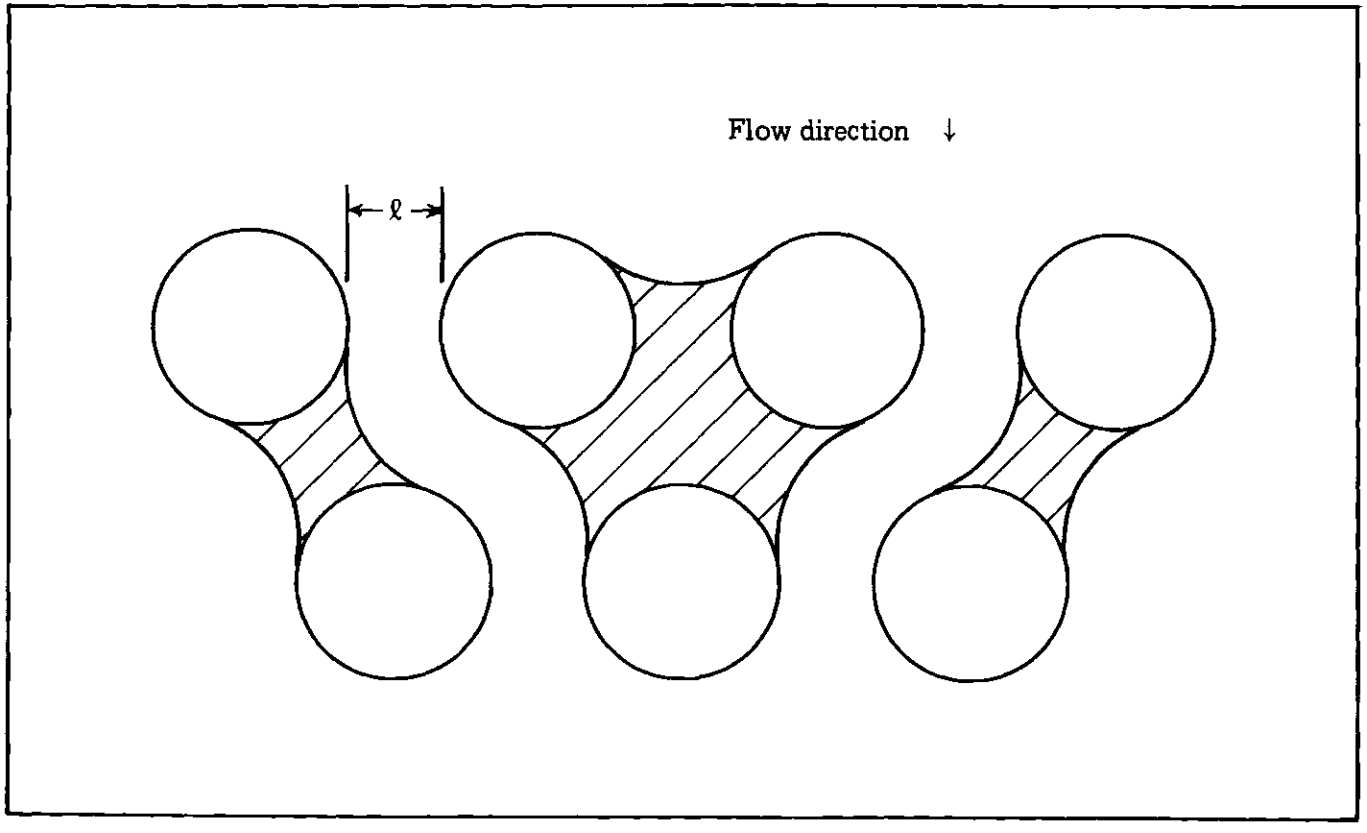

Figure 1. Definition Sketch for Channeling Model

of maintenance of some unrestricted flow channels in the porous bed throughout filtration. In this figure the circles represent sand grains and the lined areas represent matter deposited in such a manner that some passageways between grains are completely blocked and others are left completely open. Mathematically, the porous medium is represented by a number of parallel channels of uniform cross section ℓ. It is assumed that at time t, some of the channels are completely clogged while others contain no deposited material. By a straightforward mathematical derivation assuming only the validity of the Poiseuille formula, the hydraulic conductivity ratio is found to vary directly with the degree of clogging.

In a second model, the porous medium is again assumed to consist of a number of flow channels of equal uniform cross section. Deposition of fines is assumed to take place uniformly around the grains which form these channels. The heavy circles in Figure 2 represent sand grains and the light circles depict deposited material. The enlarged cross section shown clarifies the notation used with this model. In this diagram, $s/2$ is the thickness of the deposited fines around each channel, $f/2$ is the thickness of suspended fines around each channel, and $\epsilon s/2$ is the thickness of water deposited with the fines where ϵ is the void ratio of deposited fines. From this beginning, it was deduced that, for constant volumetric flow and constant ϵ, the hydraulic conductivity ratio varies proportionally with the cube of the degree of clogging.

A third model is based on the hydraulic radius, which is here

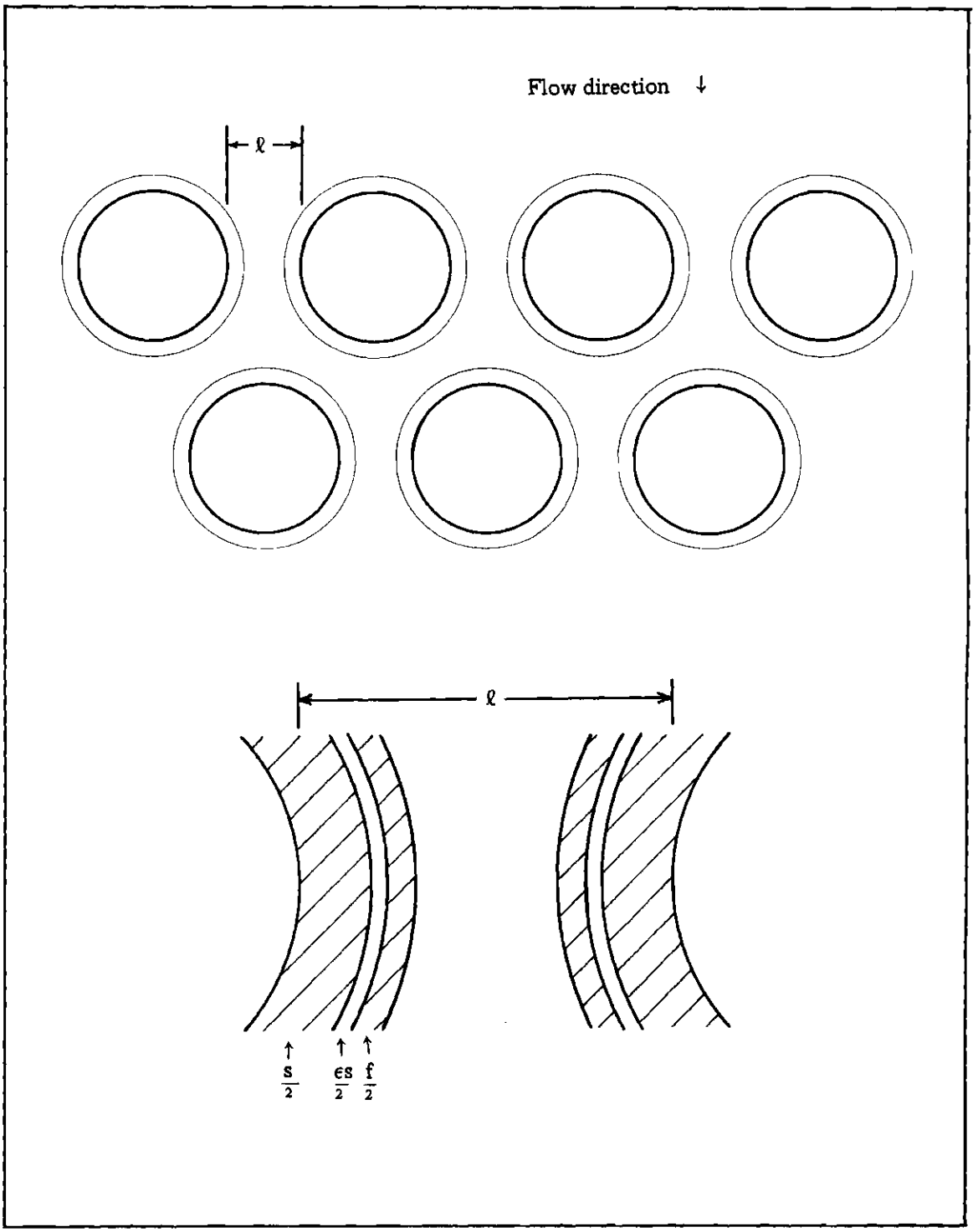

Figure 2. Definition Sketch for Uniform Deposition Model

defined as the volume of liquid in the stationary deposit divided by the wetted area of the deposited fines. The result indicates that the hydraulic conductivity ratio is proportional to

$$\frac{[n_m - (\epsilon + 1)S]^3}{[1 - n_m + (\epsilon + 1)S]^2} \qquad (1.33)$$

where n_m is the porosity of the matrix and S is the degree of clogging.

Experimental work testing the validity of these models showed a satisfactory correlation only with the hydraulic radius model, for the materials, equipment, and operating conditions involved.[61]

Possible Filter Mechanisms

It has been assumed by some authors that deposition of matter in a filter is caused by one or a combination of three mechanisms: straining, settling, and/or surface attraction. In turn, these may be brought about by forces such as gravity, inertia, van der Waals force, shear force between high and low velocity regions, thermal forces, and/or electrical forces. In the following discussion, each force or mode of action will be treated separately.

Straining

Perhaps the most obvious mechanism of removal of particulate matter from water is the deposition on the filter surface of particles too large to enter the openings in the filter. Ives and Gregory[62]

dismiss completely the idea of a sand filter as a strainer, noting that, if this mechanism is operative, a microstrainer or sieve would be more appropriate. Their idea may have merit, but the concept of a strainer cannot be passed over so lightly. In a detailed study of the filtration of non-colloidal sized particles, Sakthivadivel[61] observed "bridging" and considered it a distinct mechanism. Perhaps it could better be considered a form of straining, as both are manifestations of physical hindrance.

Even though a particle is considerably smaller than the average pore diameter in a filter, interstitial straining can occur in the narrow corners adjacent to the points of contact of the filter grains. This serves to lessen the size of the pore, decreasing the probability of passage of particles arriving later. A particle too large to pass through then forms a "bridge" or wedge against the filter grain and/or the deposited matter. It is not necessary that flow be in the downward direction, as particles will be maintained in place by hydrodynamic drag. The critical factor in this situation is the ratio of the diameter of the grains of the filter medium to that of the suspended particles (d_m/d_s).

Also involved is the porosity of the matrix, which depends on the packing, particularly in the idealized case of spheres. When the diameter of the suspended particles is equal to or greater than half the diameter of the pore opening, the filter clogs. If $d_m/d_s \leq 5$, the filter will clog regardless of porosity, which is in keeping with the findings of Patrashev.[21] If $d_m/d_s > 14$, the filter will not clog. Between these limits possible clogging depends on the porosity of the ma-

trix. Suspended particles, often referred to as "fines," will be washed out by laminar flow when $d_m/d_s > 10$ to 12. Bertram[63] and Justin[64] recommended that to avoid removal of fines from successive layers of inverted filters or drainage blankets, the ratio d_{m15}/d_{s15} should be less than 9, where the subscripts denote the 15 percent size, i.e., 15 percent smaller than and 85 percent larger than.

Another argument against the theory of straining as the principal removal factor can be developed by considering head loss through a filter. If physical hindrance were the predominant mechanism, most of the removed matter would be deposited on the surface of the sand with a very high head loss across the sand surface and very little at lower levels. This situation is not encountered in ordinary filtration situations.

Gravity Effects

According to a theory proposed by Hazen,[65] each opening in the filter medium may be considered as a tiny sedimentation basin where gravitational settling occurs because of density differences between the particle and the water. Removal of a particle by settling occurs when a particle reaches the bottom, and theoretically this will happen more rapidly if the density difference is greater or the settling distance less. Developed in detail by Camp,[66] this line of reasoning leads to the conclusion that the ideal basin would have a depth approaching zero, but with an accompanying requirement of an area approaching infinity. While this is not practical for settling basin designs, a filter may be considered a close approach to this ideal.

Hall[55] considered gravitational effects a major component of filter action and developed a differential equation for filtration based on

the sedimentation theory. He found good correlation between calculated results and data published by the ASCE Committee on Filtering Materials.

Cleasby and Baumann[67] studied the removal of calcium carbonate crystals and concluded that straining and sedimentation are the predominant removal mechanisms. This was based in part on microscopical examination of the sand after filtration, which revealed that most deposited crystals were on the upper surfaces of the sand grains.

Further evidence of a gravitational action is supplied in a later investigation by Ives and Sholji[54] who passed a suspension of kaolinite upward through a porous bed. The removed particles were found to be deposited on the tops of the grains, but not on the upstream faces.

In contradistinction to these findings, Stein[51] reported that sedimentation of suspended floc particles within a filter bed is not an important collection mechanism. He observed best removal at points of maximum velocity which is not in agreement with the sedimentation theory. It is therefore apparent that the sedimentation theory offers no explanation for the good removal of a floc with a density near that of water.

Inertial Impaction

During flow of a fluid containing particles of a density greater than that of the fluid, the motion of the particles will not be the same as the motion of the fluid as they approach an obstruction. The momentum of the particles makes them less subject to deviation from their course when the streamlines of flow diverge to pass the obstruction. As the stream passes the obstruction, some of the denser matter may be forced out of the stream and adhere to the surface. It is not likely that inertial impaction is a major removal mechanism in rapid sand fil-

tration, but it has been discussed by several authors.

Obvious factors involved include the velocity, the position of a particle within the stream, the angle of impingement, the viscosity of the fluid, and the difference in density between the fluid and the particle. Since momentum is proportional to velocity, an increase in velocity should increase removal efficiency. Under normal operating conditions, as the pores of a filter begin to clog, the velocity through them must increase in order to maintain a constant flow rate. The inertial impaction hypothesis may therefore be rejected, as the efficiency of filtration does not increase under these conditions.

Inertial impaction is an important mechanism in the removal of solids from gases, and it has been studied extensively.[68,69,70] It is not, however, of appreciable significance in sand filtration of water. Even in gases it is easily shown that high velocity turbulent streams are necessary for good efficiency, and this condition does not exist in sand filters. Cleasby and Baumann[67] have shown experimentally that, in the filtration of aqueous suspensions, the flow regime is laminar even when a filter is clogged to a considerable degree.

Additionally, it has been shown both theoretically[71] and experimentally[62] that inertial impaction is a negligible phenomenon with particles in the size and density range normally encountered. Ison's experimental work[62] involved the flow of a suspension of kaolinite upward through a porous bed. The particles removed did not attach to the bottoms of the grains in this upflow experiment but were found to deposit on the tops of the grains. This finding was interpreted as indicating a gravitational rather than inertial mechanism. It is also possible, however,

that forces other than gravity caused the observed deposition pattern. Due to streamline divergence immediately after passage through a constriction, the forward (in this case upward) motion would be at a minimum, and the particles would be subjected for a longer time to the effect of whatever forces might be operating, such as electrical interactions.

Diffusion

It is well known that matter in solution will migrate toward areas of low concentration, and the same is true of small suspended particles. This offers an explanation of particle removal through diffusion into regions within the filter where the flow is essentially zero. Experimental work has shown that, in the presence of a concentration gradient, clay particles will diffuse across streamlines in this fashion.[72] It is also possible that the concentration of particles in the stagnant region may exceed that of the stream because of the tendency of colloidal particles to accumulate in regions of low shear,[73] even if this requires migration against a concentration gradient. Shear is thus an opposing force to concentration uniformity and an equilibrium probably develops. It has been demonstrated that these forces are operative,[74,75] but it will be shown in Chapter V that diffusion forces are too small to contribute an appreciable effect.

Brownian Movement

Brownian motion is the random movement of particles suspended in a fluid as a result of random elastic collisions with atoms and molecules. Brownian movement has been discussed and rejected as a major filtration mechanism by a number of investigators. The following is a summary of their conclusions.

When the particles are very small, consisting perhaps of only a few atoms or molecules, they move in a path characterized by abrupt changes of direction. Larger particles must be struck many times before they change direction, and the path is a tortuous curve instead of a series of straight lines. An equation derived by Einstein[76] and verified experimentally by Perrin[77] for the average square of a particle displacement in time t is

$$\overline{\Delta x^2} = 2Dt \tag{1.34}$$

where D is the diffusion coefficient. $D = RT/N'K_r$ and, therefore

$$\overline{\Delta x^2} = 2RTt/N'K_r \tag{1.35}$$

where K_r is the resistance factor, R the gas constant, T the absolute temperature, and N' Avogadro's number. If the particles are large enough for Stokes' law to hold

$$\overline{\Delta x^2} = 2RTt/N'\ 3\pi\eta d \tag{1.36}$$

where η is the viscosity and d is the particle diameter. In air at 760 Torr pressure and 20°C, this displacement is about 3.7×10^{-3} cm for particles 0.1 μ in diameter and about 7.4×10^{-4} cm for particles 1.0 μ in diameter.

The diffusion coefficient is defined as the quantity of matter passing through unit surface in unit time under a unit concentration gradient and has the dimensions of length squared per unit time. Let

dc/dx represent the concentration gradient in a suspension of particles whose concentration varies with position. The rate of flow of mass of particles through a surface of area S would be

$$\frac{dm}{dt} = SD \frac{dc}{dx} \tag{1.37}$$

Values for D calculated by Fuks[53] for particles in air at atmospheric pressure and 23°C are given in Table 2. Exactly comparable data for spheres in water are not available, but measured values for some high polymers are listed in Table 3. While the exact size of these polymer molecules is subject to some uncertainty, the tobacco mosaic virus has been found to be a rod 300 mμ in length, 15 mμ in diameter. The other materials are thought to have diameters in the order of 10 to 50 mμ.

Table 2. Diffusion Constants of Spherical Particles in Dry Air at Atmospheric Pressure and 23°C

r (microns)	D (cm^2 sec^{-1})
10^{-3}	1.28×10^{-2}
10^{-2}	1.35×10^{-4}
10^{-1}	2.21×10^{-6}
1	1.27×10^{-7}
10	1.38×10^{-8}

Table 3. Average Diffusion Coefficient D ($cm^2 sec^{-1}$) of Some High Polymers

Solute	Solvent	Temperature (°C)	D x 10^7
Edestin	Water	20	3.18
Tobacco mosaic virus	Water	20	0.53
Human tubercle bacillus polysaccharide	Water	20	7.0 - 7.6
Acid treated starch	Water		11.5
Cellulose Acetate	Acetone		5.4
Polystyrene	Toluene		3.6

In the range of particles a few microns in diameter, the values in air are quite small; Brownian motion diffusion is therefore quite slow, even in a gas. Diffusion constants in a liquid are smaller, and diffusion rates correspondingly slower.

Several investigators have considered Brownian motion in this context. Stanley[50] did not give the details of his calculation but reported that it would take 20.3 seconds for a particle only 10^{-5} cm in diameter to move 10^{-3} cm due to Brownian action. During this time, roughly one half of the particles of this size would be removed. He reasoned that only a very small percentage of particles would be within this distance of a sand surface and therefore rejected this mechanism.

Brownian movement velocities were measured by a number of early investigators,[79] and a representative value is $v = 1.5 \times 10^{-6}$ cm/sec

for 20 mμ particles in water. The superficial velocity of water through a sand filter at two gal/ft^2/min is about 20 cm/min, and the conclusion of Stanley appears valid.

Ives[54] concluded that Brownian action cannot be a very important mechanism in liquid filtration of particles larger than about two microns and based this conclusion on direct observation. Grace[80] also agrees that the effects due to Brownian movement are negligible when compared with other removal mechanisms that have been proposed.

Flocculation and Random Contact

Fair and Geyer[81] regard floc formation in the filter as a major removal mechanism. Their discussion implies a consideration of only those waters that have been coagulated, but there is no *a priori* reason to eliminate a similar type of action for untreated waters.

When a coagulant is added, precipitation in dilute solution produces finely divided or colloidal suspensions. Agglomeration of these suspended particles yields settleable solids. Under quiescent conditions agglomeration occurs through collision of particles due to Brownian motion, and finely divided solids are brought into contact when more rapidly settling solids overtake those settling more slowly. Agglomeration is very slow under these conditions but can be accelerated by stirring the water. Velocity gradients are created which cause an increase in the number of collisions or contacts between particles.[82] The rate of flocculation depends on the number of contacts per unit time and increases with number and size of the particles and the available energy causing the motion. The process does not continue indefinitely; it is limited by the concentration of particles and ease with which the

flocs can be broken apart by shearing forces. The mathematical aspects of flocculation were presented by Smoluchowski.[83]

In viewing flocculation in these terms, it may be concluded that any process increasing particle collisions would consequently increase the agglomerate size. In a filter the alternate constriction and divergence of stream lines would impart lateral motion to suspended particulates and might well bring them into collision with each other or with the large surface of the grains of the filter. When large enough, the particles would be trapped by subsequently encountered constrictions between pores. Fair and Geyer claim further that the deposited material restricts the passageways, increases shearing forces, and tears the floc particles apart so that they penetrate further into the filter. This latter suggestion is similar to the theory of Mackrle.[58]

Considering deposition as a function of chance contact, Stein[51] suggested that the probability of removal of a suspended particle is directly proportional to the square of the particle diameter and inversely proportional to the cube of the diameter of the bed particles. There is nothing obviously illogical about this process and it requires only the postulation of a force to keep the particles from redispersing.

Van der Waals Forces

There are a number of known forces of attraction and repulsion at the molecular level, one of which is that deduced by van der Waals. These attractive forces vary inversely as the sixth power of their distance of separation, and thus are of considerable magnitude when the distance approaches the molecular diameter. This is the situation that exists in liquids, where these cohesive forces account for the major

properties of the liquid state. Forces of repulsion are also greater at very small distances, but in general are of a considerably smaller order of magnitude.

The cohesive forces are additive so that, for large aggregates, the force becomes inversely proportional to the cube of the separation distance. Because of the rapid decrease of effect as distance increases, this does not appear likely to be a dominant removal mechanism. It may, however, be the major force that prevents redispersion of removed matter.[84]

The van der Waals forces have been measured experimentally between polished quartz plates in vacuum.[85] Actual values of the attractive energy between a sphere (1) and an infinite plane (2) in water may be calculated from the expression $V = A_{12}a/6h$ where A_{12} is the Hamaker constant, a is the radius of the sphere, and h is the separation distance. Values of the constant are obtained from optical data for the two materials and for water. The energy[86] for most materials in water is in the range of 0.2 to 5×10^{-13} erg.

Surface Forces

According to physical chemistry theories, there are forces of attraction and repulsion at surfaces. Both decrease with distance from the surface, but at different rates. This is shown in Figure 3 where the attractive forces vary as the sixth power of distance, and repulsion varies exponentially, assuming the potential on the two surfaces to have the same sign. The combined curve has a maximum which represents an energy barrier. Any approaching particle with sufficient energy to overcome the barrier will be strongly attracted and bound to the grain

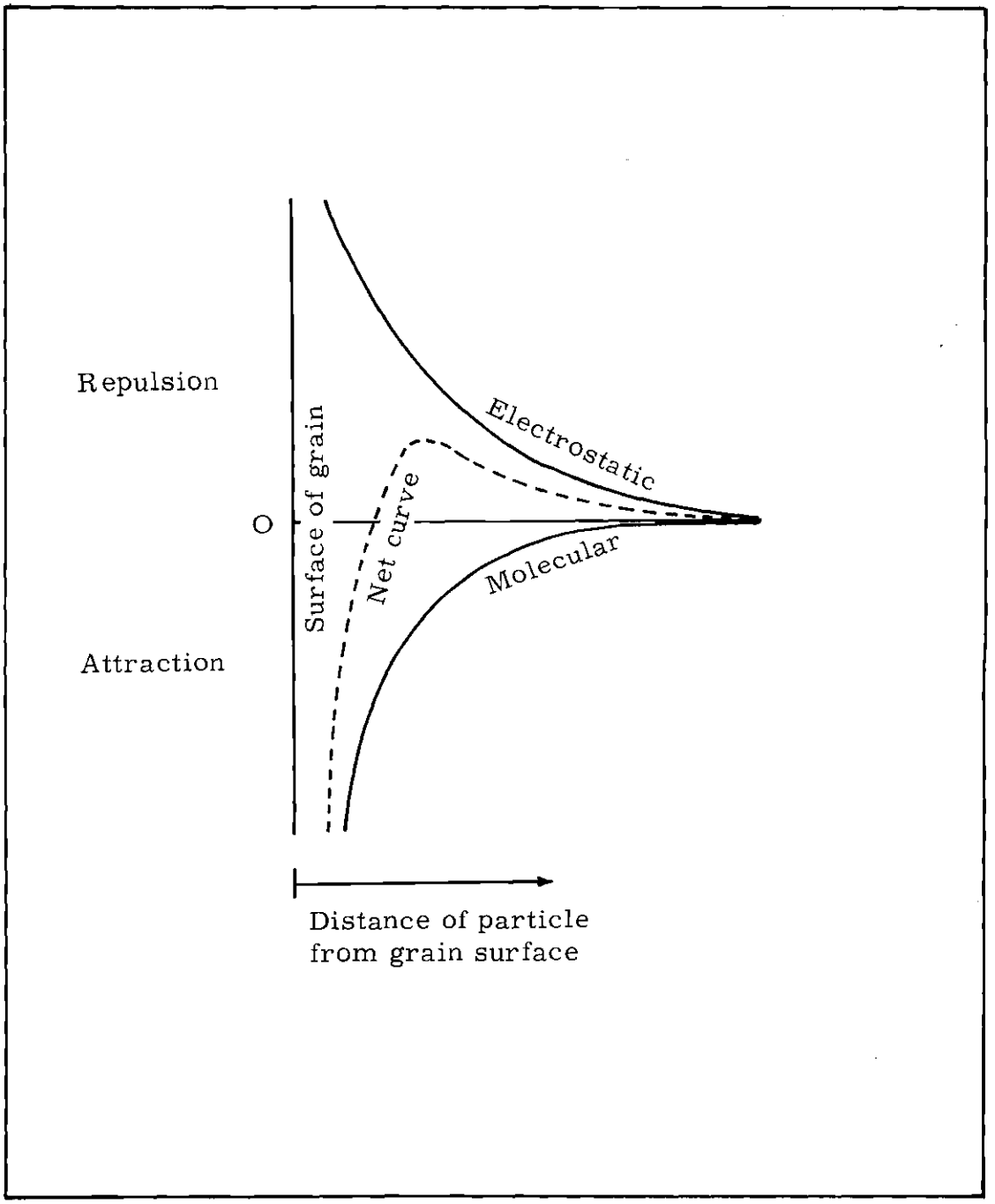

Figure 3. Variation of Forces with Separation Distance

surface. The required kinetic energy can be supplied hydrodynamically or thermally. It may also be possible to reduce the barrier by diminishing the forces of repulsion. Primary electrostatic forces of repulsion are not present when the two surfaces are oppositely charged.

The origin of the electrostatic forces[87] involved is important to a clear understanding of the resultant actions. All solid particles have a charge at their surface when placed in water due to one or more of the following:

1. ionization of molecules at the particle surface,

2. unneutralized net charges due to imperfections in the crystal lattice,

3. direct chemical reaction with specific ions in the solution with the formation of chemical bonds, and

4. weaker, physical adsorption of ions from solution due to hydrogen bonding and van der Waals forces.

The surface charge, regardless of origin, attracts ions of opposite charge and a thin tight layer is formed. This layer in turn attracts ions of opposite charge and a second, less sharply defined layer is formed. The two layers result in a zero net potential. This diffuse double layer theory was developed by Stern[88] in an attempt to explain the behavior of colloids. In filtration these surface forces are present on both the sand grains and the suspended particles. It should also be noted that the thickness of the stationary layer is not invariable. It may be increased by the addition of an electrolyte, which offers an explanation of observed effects on filtration due to added ions in solution.

Experimental evidence[89] shows that newly formed quartz surfaces are negatively charged, but the charge is reduced by iron staining. Natural particles in water such as silt, bacteria, and algae are negatively charged, whereas the flocculant hydroxides of iron and aluminum are positively charged and are more readily removed by filtration.

Mackrle,[58] however, considered that electrostatic forces were not the primary force in the case of aluminum or ferric hydroxide flocs and attributed their removal to van der Waals forces. He showed that the constant of molecular attraction between the flocs and such materials as quartz, calcium carbonate, glass, and lead when immersed in water is almost independent of the materials involved.

The work of Stanley[50] which showed the best filtration of iron floc at its isoelectric point, indicates the involvement of electrokinetic effects. Sanford and Gates[90] state that such forces do not exert a major influence in rapid sand filtration. Cleasby and Baumann[67] report electrokinetic forces predominate in the removal of hydrous ferric oxide floc. This variation of opinion is unresolved at present, but it may be that electrokinetic effects are confined to interactions between floc particles. It is also possible that the mechanism of removal of hydroxide flocs is different from that of other suspended particles.

O'Melia[91] investigated the filtration of negatively charged algae which passed easily through his experimental filters. Removal efficiency was greatly improved, however, by the addition of iron coagulant, indicating electrokinetic forces to be of primary importance. This conclusion is also supported by the increased efficiency of removal of viruses, algae, and bacteria when a coagulant is used.[92,93,94]

It is evident that much further experimental work is needed to resolve these conflicting ideas, and the present work is intended as a contribution to this end.

CHAPTER II

CURRENT FILTRATION THEORY

Based on the literature survey summarized in Chapter I, it is possible to present current theory very succinctly. Although there are some dissenters to most of the details, the concensus is as follows: certain physical mechanisms can account for the removal of some specific kinds of particles, but no single mechanism is responsible for removal of all types and sizes of suspended particles. It is generally thought that collection of colloidal particles is governed by diffusion. Hydrodynamic and interception mechanisms control larger particles in the range of perhaps one to fifty microns diameter whose density is close to that of water. Gravity is considered to control particles in the one to fifty micron range that are much denser than water. There are no very clear divisions of influence and there are overlaps in these mechanisms.

It has been demonstrated that the surface characteristics of the porous medium are one determinant of filtration efficiency, and velocity of flow through the medium is another. Water chemistry is also a demonstrable factor. A number of mathematical models have been found useful in describing filtration and the effects of certain variables. Application of these models has been limited by the necessity for evaluating empirical constants, and this can be done only by experimentation.

There is unanimous agreement that the process is dependent on

many variables and the action observed probably comes about through a combination of different mechanisms. Using the filter coefficient, λ, as a measure of filter operation, it may be stated that

$$\lambda = Af_1 + Bf_2 + Cf_3 + \ldots + Xf_X \qquad (2.1)$$

where f_1, f_2, f_3, ... represent the filtration result brought about by each mechanism and A, B, C, ... indicate the fraction of the total effect produced by the associated mechanism. It is further surmised that, as conditions are changed, the values of A, B, C, ... may also change, so that the relative importance of each mechanism is variable. Any mechanism may be pictured as occurring in two separate steps: 1) transport of the suspended particle to a surface, and 2) attachment or immobilization of the particle at the surface. These factors seem independent, as attachment might be brought about by a given means regardless of the manner in which the particle is brought to the location where immobilization occurs.

One possible mechanism is interstitial sieving, which would occur in the crevices adjacent to the points of contact of the sand grains as illustrated in Figure 4. Any particle contained in the flow passing through triangle XYZ would be trapped if its smallest dimension were larger than length YZ. As the sides of the triangle converge at point X, by suitable placement of line segment YZ it is possible to define a triangle with a maximum width smaller than the size of any suspended particle. The portion of the flow that passes through any such triangle will, therefore, be stripped of all particles larger than the YZ dimen-

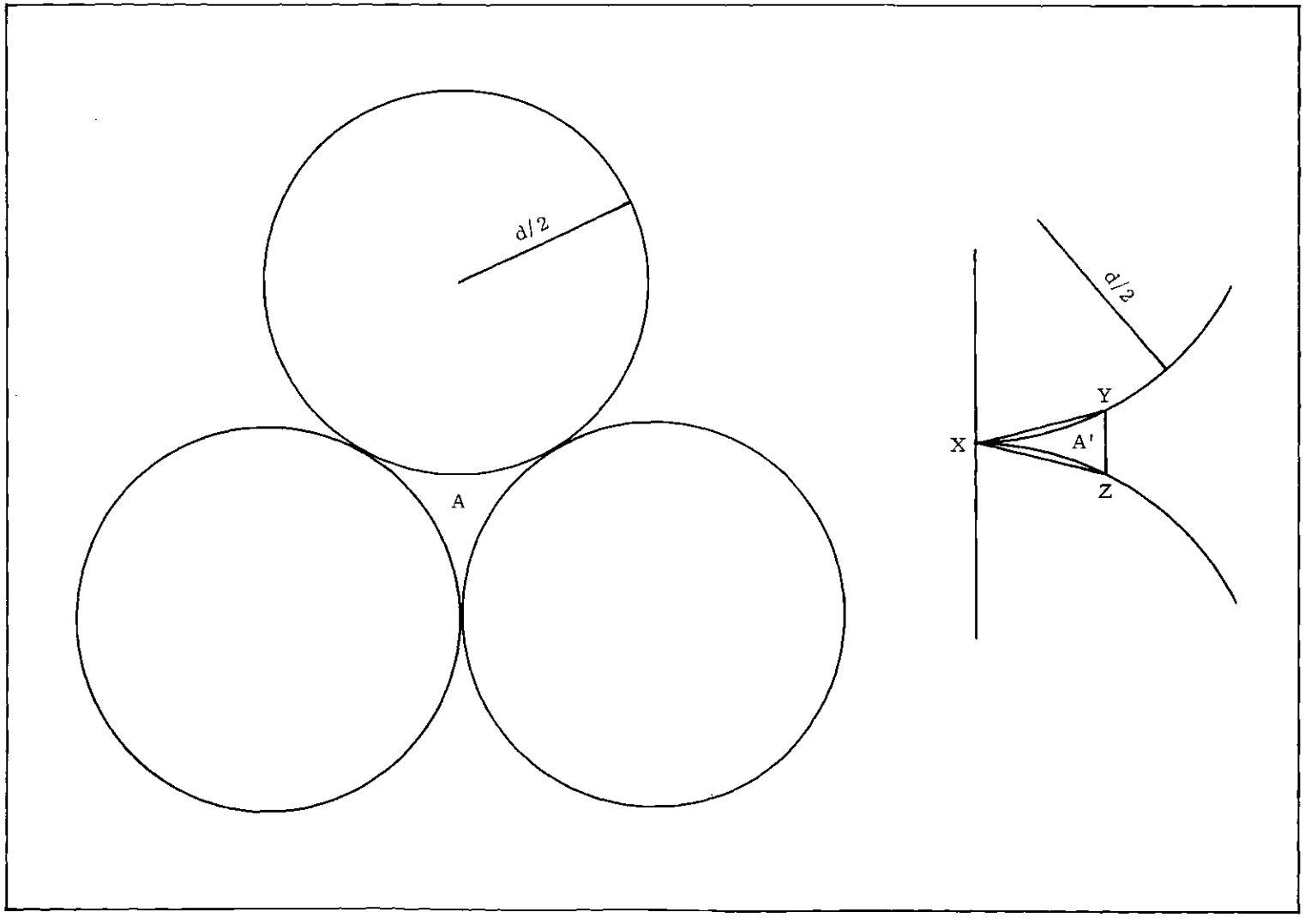

Figure 4. Idealized Geometry of a Cross Section of a Pore Space

sion of the triangle. The amount of removal at each sand layer for a given particle size is therefore determined by the ratio of the flow within the triangle to the total flow through the construction.

Viewed as a statistical phenomenon, the hindrance mechanism assumes that removal is dependent on the probability of a given particle encountering an opening through which it cannot pass. This encounter can take place if there is an opening of suitable size and the particle is in that portion of the flow passing through the opening. Based on the idealized geometry of spheres, it is assumed that every crevice in the filter is capable of trapping a particle of any size. Entrapment then may be considered as a function of the probability of the particle being located in an appropriate position in the flow stream.

The details of this mechanical sieving action were set forth originally by Hall,[55] who derived the following theoretical relationship which is most easily explained by reference to Figure 4. Assuming that the area between the arc segments is the same as that between the chords, the area A' of the constriction with a dimension less than YZ is approximately

$$A' = \frac{3}{2\sqrt{2}} D^{\frac{3}{2}} d^{\frac{1}{2}} \qquad (2.2)$$

where $D = YZ$ and is the diameter of the particle that will be trapped.

If d is the diameter of the sand grain, the total area of the constriction is

$$A = 0.040\ d^2 \qquad (2.3)$$

If interstitial straining is the only mechanism or even the predominating mechanism, the filter coefficient should be proportional to the ratio of these areas

$$\lambda \propto \frac{A'}{A} = \frac{\frac{3}{2\sqrt{2}} D^{\frac{3}{2}} d^{\frac{1}{2}}}{0.040\ d^2} \propto \left(\frac{D}{d}\right)^{\frac{3}{2}} \qquad (2.4)$$

If λ is plotted against $(D/d)^{\frac{3}{2}}$, a straight line would indicate that the two parameters are proportional to each other, and this would support the validity of the mathematical model.

In this presentation of the sieving hypothesis there seems to be a tacit assumption that the liquid and the suspended particles it contains are moving downward through the filter at a rate that is everywhere uniform, whereas this is not the situation that exists. Superficial or overall velocity is a well known variable in filtration efficiency, but the most important aspect of velocity effect may lie in the distribution of velocities across bed openings.

Among investigators in rapid sand filtration, the assumption of laminar flow is generally accepted. It should be recognized, however, that the nature of the porous medium precludes the possibility of straight or gradually curved streamlines of great length as might be found, for instance, in a pipe. The tortuosity of the flow paths presents a situation that does not closely fit the classical concepts of either laminar or turbulent flow.

One criterion for evaluation of flow regime has been established through an analysis of the relationship of head loss to bed depth. A linear relationship is predicted by Darcy's law and is found in most

porous media at low flow rates. Under conditions of turbulent flow, Darcy's law is not valid; instead of being linear, the head loss varies as a higher power of the flow rate. This may be expressed in an equation of the form $h_L = f_f \frac{\ell}{d} \frac{V^N}{2g}$ where h_L is head loss, f_f is a proportionality coefficient called the friction factor, ℓ and d are conduit length and diameter, g is the gravitational constant, V is velocity, and N is a constant usually taken as 1.8 or 2.0.[95] An early conclusion was that turbulent flow begins at the upper limit of Darcy's law adherence, but experimental evidence indicates otherwise.

In porous media the Reynolds number is calculated from the expression $R_e = Vd/\nu$ where V is velocity, d is the diameter of the grains of the porous medium, and ν is the kinematic viscosity. Using typical values of flow rate and sand size, the Reynolds numbers associated with rapid sand filtration have a value no greater than one or two.

According to the experimental data of several investigators,[96,97,98,99,100] these Reynolds numbers fall in the realm of laminar flow, based on adherence to Darcy's law, but relatively close to the transition range (starting around 10) where head loss is no longer directly proportional to depth.

Schneebeli[101] visually observed the onset of turbulence at a Reynolds number of about 60, whereas Hubbert[102] found it in the range of 600-700. Although there is a wide discrepancy between these observations, there is a strong suggestion that turbulence does not begin until attainment of a Reynolds number many times greater than that of the upper limit of Darcy's law. This implies that the Reynolds number is not a good criterion for evaluating the beginning of turbulence in flow through

porous media, and also that laminar flows that do not obey Darcy's law may exist in porous media. Each globule of fluid moving through a porous medium follows a continuously curvilinear path at a continuously varying speed, and also with a continuously varying acceleration. At a relatively slow rate of flow (near Reynolds numbers one to ten), the inertial forces in this essentially nonuniform flow become significant. Darcy's law governs flow only when resistive forces predominate; it does not apply when the inertial forces approach the same order of magnitude as the resistive forces.

It therefore follows that, if inertial forces pertaining only to the fluid are near the magnitude required to cause deviation from Darcy's law, entrained particles denser than the fluid would be subjected to even greater inertial forces. In passing through a series of zones of alternately diverging and converging streamlines, there is considerable opportunity for a particle to be transported across streamlines; it appears unlikely that a particle would remain in the same streamline through a very great depth of porous medium. In the complex geometry of a porous medium there is also the likelihood of eddies produced by rapid changes in velocity due to sudden passage from a constriction into a larger space. These hydrodynamic-inertial forces may bring particles close enough to surfaces to allow other forces to capture them.

Another postulated particle collection mechanism for rapid sand filtration is gravitational sedimentation. Consider an array of spheres past which a suspension of uniform particles is flowing in the direction parallel to the acceleration of gravity. As the particle has a greater density than water, an unbalanced force will exist until the particle

accelerates relative to the fluid to the terminal velocity where the viscous drag is equal to the unbalanced gravitational force. An unbalanced inertial force also exists due to the curvatures in the streamlines passing around a sphere. However, since the acceleration of gravity is very much greater than the acceleration due to flow path curvatures, the relative displacement of a suspended particle due to flow curvatures may be neglected, and the suspended particles may be considered to be moving downward everywhere in the filter. The velocity relative to the fluid may be calculated by Stokes' law

$$V = \frac{2\ (\gamma - \gamma_0)\ D^2}{9\mu} \tag{2.5}$$

where γ is the specific weight of the particle, γ_0 is the specific weight of the fluid, D is the diameter of the particle, and μ is the absolute (or dynamic) viscosity of the fluid.

The moving particles should strike the sand grains with velocity V which is the velocity of the particle relative to the fluid anywhere in the fluid. It is assumed that the colliding particles adhere to the sand and do not reenter the fluid. Particles are brought into a region at a rate $\overline{v}C$ where $\overline{v}$ is the mean velocity of flow and C is the concentration of particles. The rate at which the particles settle on the sand grains is VC. Hence, the fraction removed in the region under consideration is the ratio of the relative velocity, V, to the mean velocity $\overline{v}$. The rate factor for the gravitational theory is therefore

$$\lambda \propto \frac{VC}{\overline{v}C} \quad \text{or} \quad \lambda \propto \frac{V}{\overline{v}} \tag{2.6}$$

and

$$\lambda = K\ D^2/\bar{v} \qquad (2.7)$$

where K is a grouping of the several constant coefficients. From this expression, it follows that a plot of λ versus D^2 at constant velocity $\bar{v}$ should produce a straight line.

For particles of small size, other forces or mechanisms must be invoked to explain the usual results of sand filtration. The most likely explanation seems to lie in the realm of chemical or physico-chemical effects. The work of O'Melia[91] and Crapps[103] clearly demonstrates that filtration is influenced by the presence of different dissolved ions, a circumstance which does not affect the geometrical structure of either the medium or the suspended particles and should, therefore, be without effect on a purely mechanical process.

Forces other than strictly mechanical ones that have been mentioned in the literature include electrostatic repulsion, van der Waals forces, solvation, hydrogen bonding, coordinate chemical bond formation, ion exchange, and electrokinetic phenomena. The general designation "surface forces" is widely used in discussing the overall effect. All of these individual forces have been shown to perform in a manner that would explain particle attachment, at least under certain specific conditions. The prevalence of any one of these forces or a specific combination of them as the dominant cause of particle attachment has not been shown.

Of these various forces, the only ones of appropriate magnitude are the van der Waals force and the Coulomb forces associated with elec-

tric double layers present at liquid-solid interfaces. It is usually presumed that attachment of small particles occurs by the van der Waals attractive force, aided or opposed by Coulomb forces of attraction or repulsion, depending on the signs of the charges on the surfaces involved. It is logical to assume that repulsion is more likely because, after filtration has proceeded for a time, the grains of the medium become coated in large measure with captured particles; subsequent interactions must then be between the double layers of the captured and the mobile suspended particulates.

The magnitude of some of the surface and hydrodynamic forces can be calculated from theoretical considerations and verified at least to some extent by experimental evidence. There is considerable evidence from the field of colloid science for interactions due to electrical double layers, and the results are applicable to the somewhat larger particles typical of rapid sand filtration.

At the interface between a solid and an electrolyte solution, a double layer is conceived to be present, composed of the charged surface and a diffuse, enveloping "ion atmosphere" in which ions of charge opposite to that of the surface are most concentrated near the surface and thin out as distance from the surface increases. Closest to the surface is a fixed layer of almost immobile ions (the Stern layer) across which there is a sharp potential drop; the potential decreases more gradually across the increasingly more diffuse region and reaches zero potential in the bulk of the solution. This is represented in Figure 5. The potential ψ_δ, just outside the fixed layer depends largely on the ionic strength of the solution as this will determine the electrical capacity

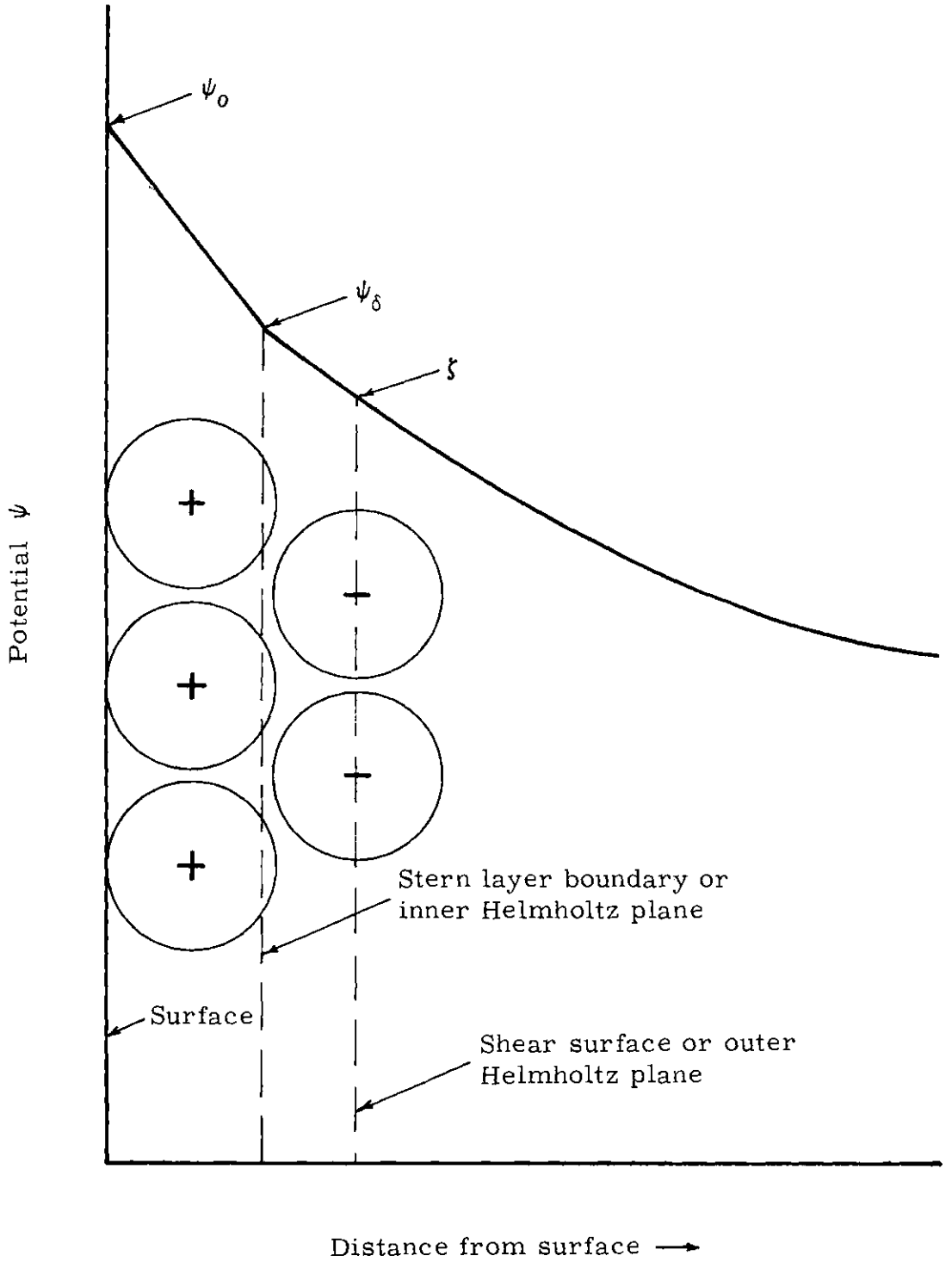

Figure 5. Diagram of the Electrical Double Layer

of the diffuse layer, and this will in turn determine how the potential drop from ψ_o to 0 is divided between the two layers.

From consideration of the relationships between charge density and potential in the double layer

$$\psi = \psi_o e^{-\xi x} \tag{2.8}$$

if $Z\psi_\delta \ll 25$ mV

where ψ = the potential at distance x from the fixed layer

Z = the valence of the ions in the double layer

ξ = the reciprocal of the double layer thickness and is equal to

$$\left(\frac{4\pi e^2 \Sigma c_i Z_i^2}{\epsilon kT}\right)^{\frac{1}{2}} \tag{2.9}$$

where e = the charge on the electron

c_i = the concentration of ions of valence Z (number per cc)

ϵ = the dielectric constant of the liquid

k = Boltzmann's constant

T = absolute temperature

When two surfaces approach one another, the double layers overlap, producing a potential distribution between them, as shown in Figure 6, if the potentials ψ_o are equal, or as shown in Figure 7 if they are not equal.

The particles to be filtered from water are sufficiently small in comparison with the filter grains that the surface force relationships may be approximated by a small sphere approaching an infinite plane.

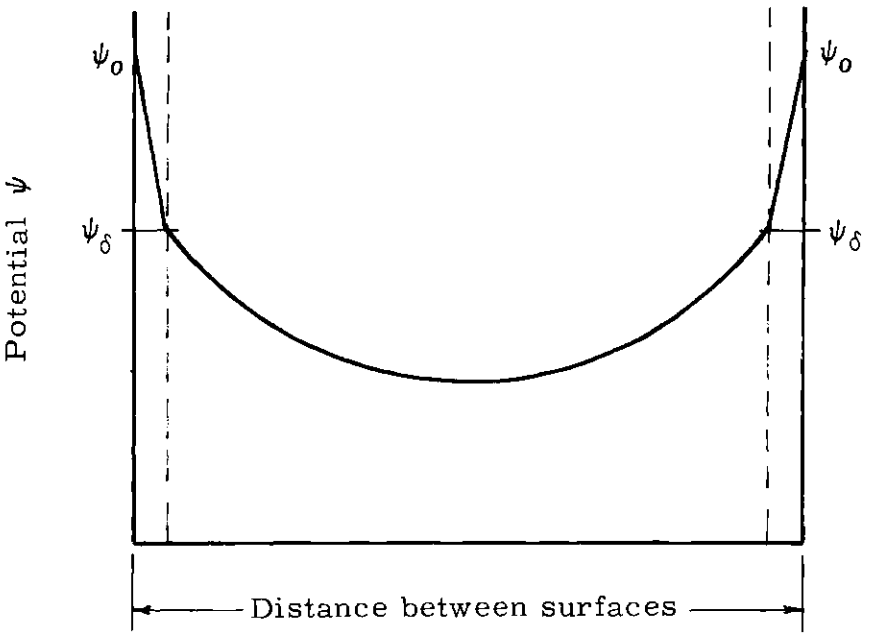

Figure 6. Distribution of Potential between Two Close Surfaces with Equal ψ_σ Potentials

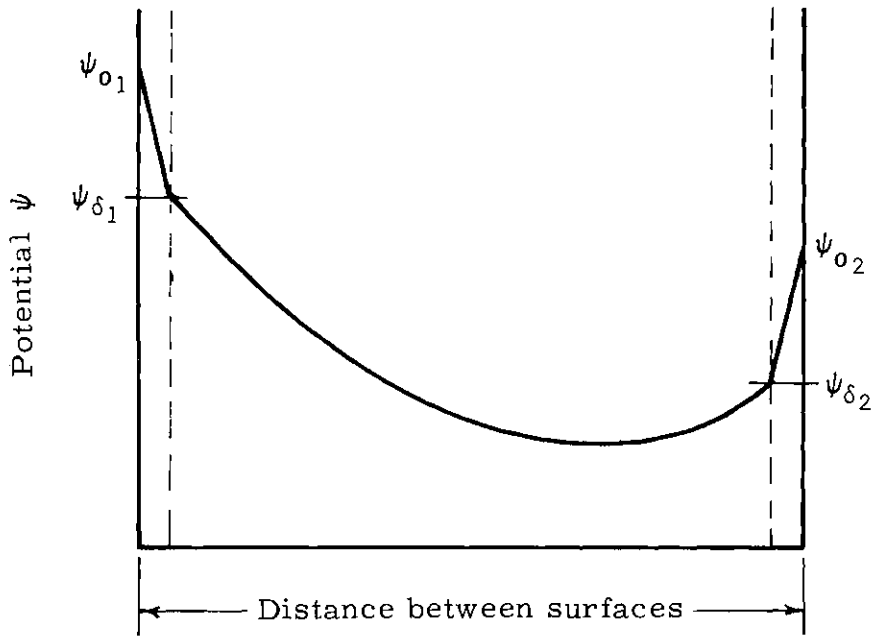

Figure 7. Distribution of Potential between Two Close Surfaces with Unequal ψ_σ Potentials

Based on this assumption, it has been shown[84] that the interaction energy is

$$\frac{V_r}{a} = \frac{128\pi\ kTc}{\xi^2}\ \gamma_1\gamma_2\ \ln\ (1 + e^{-\xi y}) \qquad (2.10)$$

where V_r = the double layer interaction energy

a = the sphere radius

c = concentration of ions of one type per cc

y = sphere-plane separation

and

$$\gamma = \frac{\exp\ (Z\ e\ \psi_\delta/2kT) - 1}{\exp\ (Z\ e\ \psi_\delta/2kT) + 1} \qquad (2.11)$$

Subscripts 1 and 2 refer to sphere and plane, respectively. Substituting and inserting numerical values pertaining to water at 25°C, the equation becomes

$$\frac{V_r}{a} = 9.24 \times 10^{-6}\ \frac{\gamma_1\gamma_2}{Z^2}\ \ln\ (1 + e^{-\xi y}) \qquad (2.12)$$

For purposes of the present discussion, forces instead of energies are required. Differentiation of the energy equation with respect to separation distance, y, give the equation

$$F_r = 9.24 \times 10^{-6}\ \frac{a\ \gamma_1\gamma_2}{Z^2}\ \frac{\xi e^{-\xi y}}{1 + e^{-\xi y}} \qquad (2.13)$$

This may be used in calculations of double layer interactions in filter

systems.

The attractive forces between atoms and molecules, arising from induced or permanent dipoles, are known as van der Waals forces. The energy of attraction varies inversely as the sixth power of the separation distance and may be calculated when values of the constants are available.

Hamaker,[86] noting that the total interaction between two aggregates can be determined by adding the contributions of all interacting pairs, showed that the mutual energy of two spheres of diameter D_1 and D_2, a distance y apart and containing q atoms per cm^3 which interact with an energy μ_1/r^6 is

$$E = -A_Q(x) = -\frac{A}{12}\left\{\frac{Q}{x^2 + xQ + x} + \frac{Q}{x^2 + xQ + x + Q} + 2\ln\frac{x^2 + xQ + x}{x^2 + xQ + x + Q}\right\} \tag{2.14}$$

where $A = \pi^2 q^2 \mu_1$ $\qquad x = \frac{y}{D_1}$ $\qquad Q = \frac{D_2}{D_1}$

μ_1 = the London-van der Waals constant

r = the separation distance of the interacting pairs

When x is much less than 1, this equation is approximated by

$$E = -\frac{A}{12}\frac{Q}{x(Q + 1)} \tag{2.15}$$

Let D_1 represent the diameter of the smaller sphere. A limit is then reached when $D_2 \to \infty$ and $Q \to \infty$, which is the case of a sphere and an infinite mass bounded by a flat surface. The corresponding equations are

$$E_{\infty}(x) = -\frac{A}{12}\left(\frac{1}{x} + \frac{1}{x+1} + 2\ln\frac{x}{x+1}\right) \qquad (2.16)$$

and

$$E_{\infty}(x) = -\frac{A}{12x} \quad \text{when } x \ll 1 \qquad (2.17)$$

Formulas for the calculation of forces can be obtained from the energy expressions by differentiation with respect to y. For the sphere and flat surface combination, we get

$$F_{\infty}(x) = \frac{A}{12}\left(\frac{2}{x} - \frac{1}{x^2} - \frac{2}{x+1} - \frac{1}{(x+1)^2}\right) \qquad (2.18)$$

and

$$F_{\infty}(x) = -\frac{A}{12x^2} \qquad (2.19)$$

Schenkel and Kitchener[104] showed that the range of separation over which these equations apply is limited. Equations 2.17 and 2.19 are satisfactory for "close" approach, but are not valid when the distance is comparable or larger than 0.1 λ_1, where λ_1 is the wavelength of the intrinsic electronic oscillations of the atoms. At distances greater than 2 λ_1, equations 2.16 and 2.18 are acceptable but there is no simple expression for intermediate distances. Another equation, with an estimated accuracy of $\pm$ 5 percent was proposed[104] for this intermediate range

$$V = \frac{Aa}{\pi}\left\{\frac{2.45\,\lambda_1}{120\,y^2} - \frac{\lambda_1^2}{1045\,y^3} + \frac{\lambda_1^3}{5.62 \times 10^4\,y^4}\right\} \qquad (2.20)$$

Differentation with respect to distance y gives an expression in terms

of force

$$F = \frac{Aa}{\pi}\left\{-\frac{4.9\ \lambda_1}{120\ y^2} + \frac{3\ \lambda_1^2}{1045\ y^3} - \frac{4\ \lambda_1^3}{5.62 \times 10^4\ y^5}\right\} \qquad (2.21)$$

This equation was not used in the present work as the simpler expressions are sufficiently accurate for this discussion. A much greater source of doubt arises from the uncertainty concerning the value of the Hamaker constant A. Different authors[84,86,104] have assigned values on the order of 10^{-12} and 10^{-14} erg, so that a possible 15 to 20 percent error at certain ranges is not particularly significant.

Lifshits[105] and Dzyaloshinskii[106] devised an alternate calculation which takes into account the presence of a liquid between the surfaces. This method deals with the macroscopic properties of the materials and for sphere-flat plate interactions is

$$\frac{Va}{a} = -\frac{kV\pi^2}{4120\ n_o d^2}\left(\frac{n_1^2 - n_o^2}{n_1^2 + n_o^2}\right) \qquad (2.22)$$

where h = Planck's constant

V = velocity of light

n_1 = the refractive index of the solid materials

n_o = the refractive index of water

d = separation between sphere and plate

This equation is of theoretical interest only as the optical data for n_1 were not available and consequently the interaction energies cannot be calculated.

It is generally agreed that particle removal is a first order reaction; i.e., the removal of suspended particles from the flow is proportional to the number present in the flow. Mathematically

$$- \frac{\partial C}{\partial L} = \lambda C \qquad (2.23)$$

which states that the decrease in concentration C with depth L is proportional to the concentration. The proportionality constant λ is termed the filter coefficient. Integration of this equation gives

$$C = C_o e^{-\lambda L} \qquad (2.24)$$

where C is the concentration at depth L, and C_o is the concentration of the influent at $L = 0$. If λ is constant, the pattern of distribution through a filter bed should be represented by a decreasing exponential function. It has been found, however, that the presence of deposited material may cause a small change in λ. The fundamental equation would therefore strictly apply only when conditions are completely homogeneous throughout the bed. In reality, uniform conditions would be approached only with a completely homogeneous bed at the beginning of a filter run when no deposited matter is present or after a greatly extended run when the bed is completely saturated with deposited matter. Under the latter condition, no further filtration could occur and λ would become zero.

The purpose of this investigation was to determine the validity of certain proposed mechanisms and its application to water decontamination. Evidence concerning interstitial sieving, gravitational effects, surface forces, and pattern of deposition is presented.

CHAPTER III

METHOD, EQUIPMENT, AND PROCEDURE

Choice of Method

When a liquid containing suspended particles is passed through a sand filter, some of the particles may be retained in the sand. The spatial distribution of these particles is not always constant but depends on many factors including such variables as sand grain size, viscosity of the liquid, and flow rate. Also involved are characteristics of the suspended particle such as density, size, and size distribution. Of prime importance, however, are the relative sizes of the suspended particles and the interstices of the sand. The type of suspended matter normally encountered in commercial filtration consists of particles in a wide range of sizes. When such suspensions are filtered, it is found that the concentration of solids trapped in the sand decreases with sand depth in a generally exponential fashion. From the work of previous investigators[23,50,51,59] it is not clear whether this exponential removal should be regarded as a general phenomenon or whether it is limited to certain size ranges of particles. There is an obvious upper limit to the size of particles which can be removed in an exponential fashion, set by the size of the openings in the sand. Particles larger than the openings will be retained on the surface due to mechanical interference, while particles small enough to pass through the sand interstices may be expected to exhibit a definite vertical pattern of deposition. The pur-

pose of this investigation was to probe the relationship between particle size and deposition pattern under different conditions, and it was therefore necessary to determine the concentration of deposited matter in different vertical layers of the filter.

The chosen method of study utilized a model filter constructed to simulate a full-scale commercial filter of the type used in municipal water treatment plants. The suspension consisted of sized fractions of ground vermiculite particles in tap water. These particles were labeled with a radioisotope so that after deposition in the sand their position could be determined with radiation detection equipment.

The two major problems facing any investigator in this field are 1) the difficulty of preparing a reproducible suspension of particles of desired size and 2) determining rapidly and reliably the concentration of trapped matter at different depths in the filter. These problems have been dealt with in a number of ways by previous investigators. Particle size has been controlled by using unisize plastic particles, by using a naturally uniform material such as algae of a certain species, or by preparing a floc under very carefully controlled conditions. The concentration of trapped material has been determined by measurement of head loss through a filter bed by use of piezometers at different depths in the filter, by removing samples of the bed for individual analysis, and by detecting matter previously labeled with a radioisotope. A routine measure of commercial filter operation is determination of the turbidity of the effluent, but this method was not applicable in the present case. The amount of added solid particles was so small that the turbidity (if any) of the water passing through the filter was undetectable with a

standard Hellige turbidimeter. Such a simple evaluation would be highly desirable, particularly in situations involving continuous feed of suspended matter.

For those with suitable facilities for handling radioisotopes, the radiotracer method of study offers distinct advantages. It eliminates the need for disturbing the filter matrix, and therefore many determinations can be made during a single run. With a well-collimated detector and a suitable level of activity, statistically reliable data can be rapidly accumulated. Constant vigilance is required for the safe handling of radioisotopes, but the small possible hazard to personnel is far outweighed by the advantages. Consequently, this method was chosen for the present work.

Previous investigators using this method have typically used plastic tubes of about two inch diameter to hold the filter sand and have mounted the detector outside of and adjacent to the tube, but for the present investigation, a larger filter area was selected to minimize wall effects and to allow the filter to be backwashed in the same manner as a full-scale filter. This arrangement facilitated thorough backwashing and compaction in a reproducible manner. More importantly, there was room to install the vertical plastic tube holding the collimated Geiger tube detector inside the filter box and down through the filter medium without affecting filter operations. In this manner the detector was surrounded with activity, a more favorable (near 4π) counting geometry than locating the detector in an external position.

The Geiger tube detector was located inside a cylindrical shield containing a horizontal slit (see Figure 13). The collimation so pro-

vided is essential to restrict the scan of the detector to a thin horizontal layer of the filter. The depth of this layer "viewed" by the detector can be adjusted by changing the width of the slit. From the standpoint of the data required, it was desirable to make the slit as thin as possible, but as the slit width is decreased, the observed count rate is also lowered. But high count rates are needed because of statistical considerations, and this demands a higher level of activity in the filter layer under observation. Very high levels of activity, while furnishing good statistics, would become unduly hazardous to the investigator, so it was necessary to reach a suitable balance between slit width and activity level.

Factors Investigated

As outlined earlier, the assumption of most investigators has been that removal of suspended matter follows an exponential law, that is, $C = C_o e^{-\lambda L}$, where C_o is the initial concentration of deposited matter at $L = 0$, and C is the concentration at depth L, and λ is the filter coefficient. Size of suspended particle does not appear in this equation but is involved because λ is a function of particle size. It was the purpose of this investigation to determine if filtration follows an exponential law for suspended particles of narrow size range under conditions where λ is constant. Another purpose of this investigation was to determine the relationship of λ to particle size and/or size of pores of the filter matrix.

The factors investigated included size of suspended particles, size of bed grains, size of bed pores, bed medium, and variation of

deposition pattern with time. Ten different particle size fractions and three different sand sizes were studied. The use of an anthracite bed provided a change in filter medium to contrast with the sand which was used in the majority of the experiments.

Description of Equipment

The experimental equipment was designed so that the desired information could be obtained as expeditiously as possible. This required a container that would hold the sand bed in such a manner as to allow downward filtration flow at rates up to about six gal/ft^2/min and upward flow for backwashing to the capacity of the water supply. A radiation detector and a means of positioning it reproducibly at various elevations within the filter were also needed. In order to minimize experimental error, effort was exerted to provide for all operations to be performed in a reproducible manner. It was decided that these goals could best be achieved by placing the detector inside a filter of a design approximating that of a municipal rapid sand filter in its vertical characteristics, and the result was considered to be representative of a large filter bed.

The equipment consisted of two eight-foot high filter boxes slightly less than one square foot in area made of plywood and which contained the porous sand bed which was about 18 inches deep. In each filter box was placed a vertical tube inside which a shielded and collimated Geiger tube could be moved up and down. Suitable plumbing and electronic counting equipment completed the apparatus. Pictures of the filters and auxiliary equipment are shown in Figures 8 and 9 and dimen-

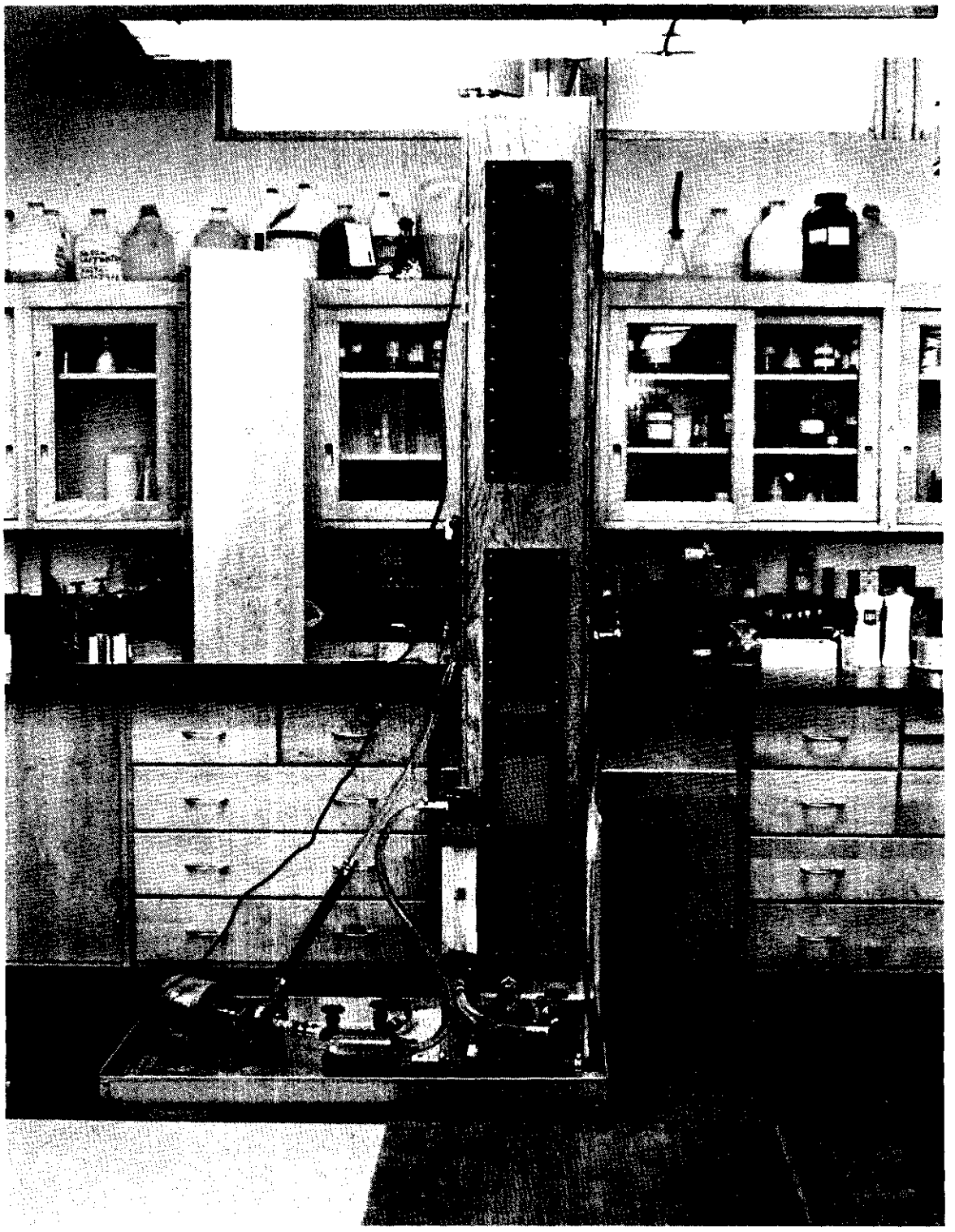

Figure 8. View of Filter No. 1

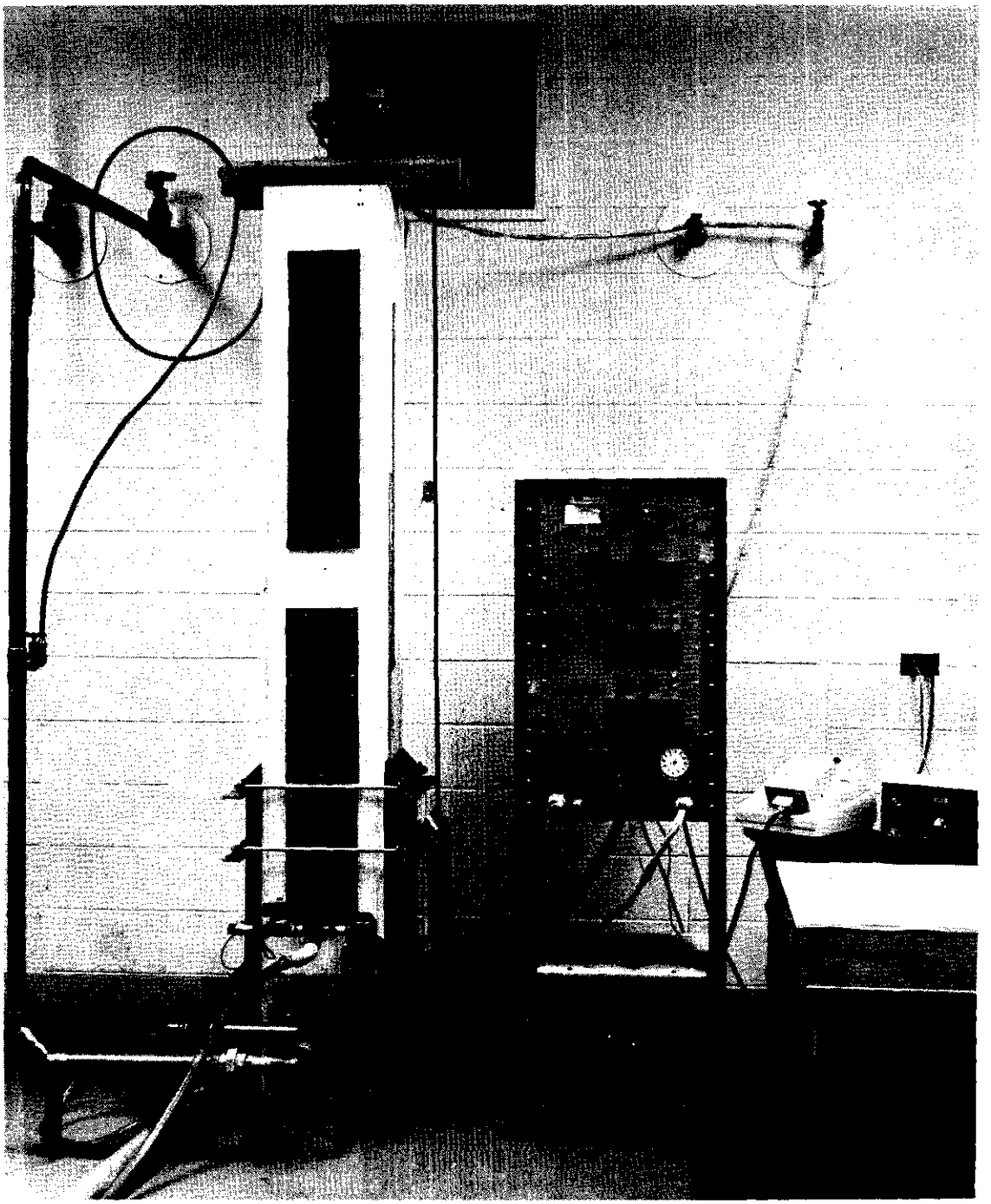

Figure 9. View of Filter No. 2 and Automatic Counting System

sions are included in the drawing, Figure 10. The first filter was installed in the Dangerous Reactions Laboratory of the Chemical Engineering-Ceramic Engineering Building on the campus of the Georgia Institute of Technology; the second filter was placed in the Unit Operations Laboratory of the same building.

The structural material was three-fourths inch marine plywood; lengths of fir two-by-fours added at the bottom increased rigidity and stability. To avoid leaks, all parts were carefully fitted and securely fastened. Butyl rubber caulking compound was used in all joints and found to be quite effective as a sealant. Channeling of flow between the porous bed and the container wall was obviated by a lining of one-fourth inch foamed neoprene. The first filter box was lined throughout, but in the second filter box only the section encompassing the bed was lined. In the first instance, the lining was attached to the bare wood; the second filter was painted inside and out with a chemical and water resistant coating before the lining was applied. The coating, which was white, increased interior visibility markedly. The water-tightness of filter #2 was later increased by the addition of heavy angle iron clamps as may be seen in Figure 9.

Both the bed and liquid above it were visible through large Plexiglas windows which were gasketed to the outside of the box with foamed neoprene and held in place with bolts and wing nuts. These readily removable windows permitted inspection of the bed and allowed visual alignment of the shield slit with the sand surface. The bolt heads were countersunk on the inside and then covered with caulking. This arrangement proved water tight throughout the study.

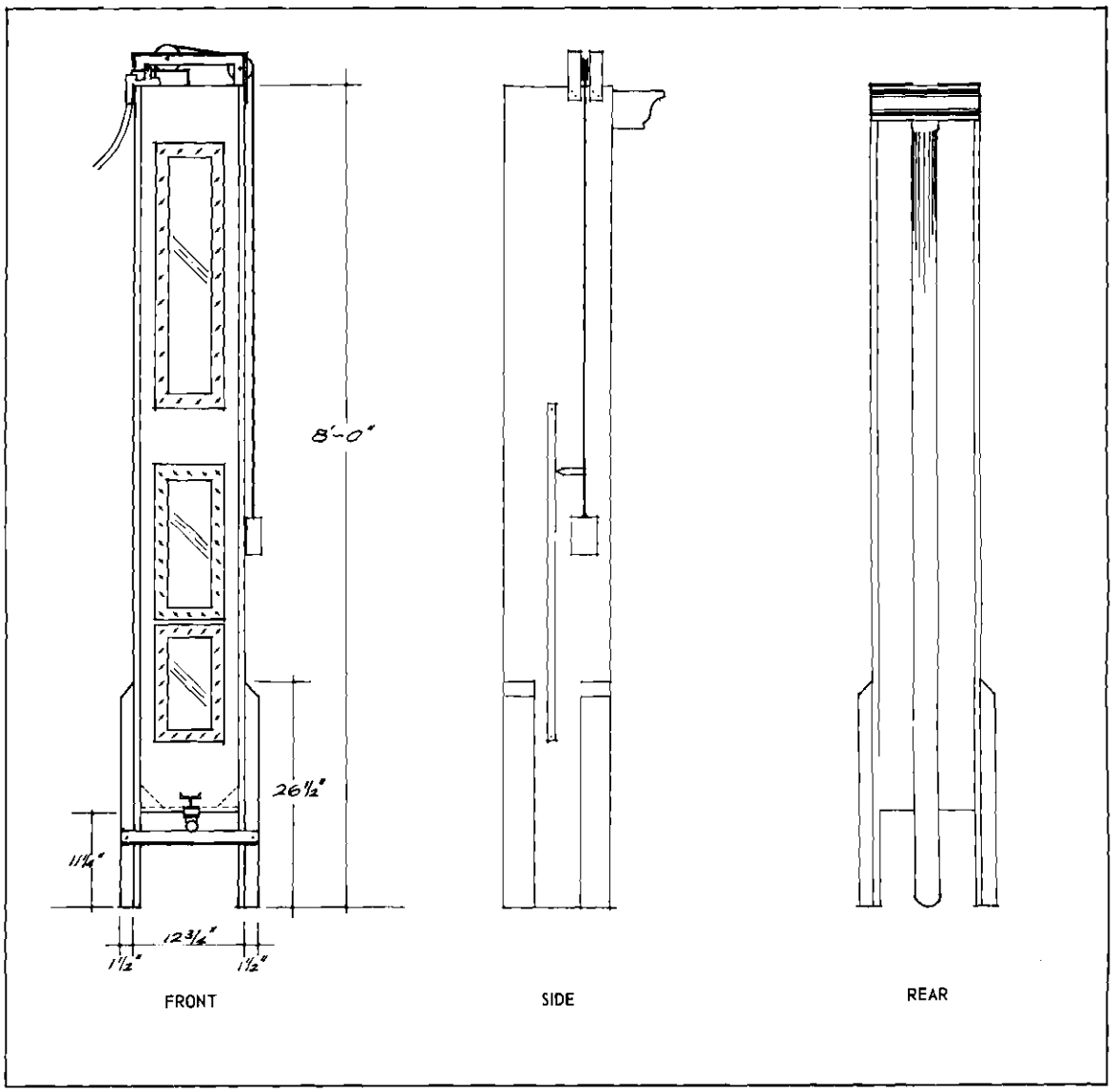

Figure 10. Details of Filter Construction

Provision was made for water flow in both a downward and upward direction, either once through or in circulation. A constant head of about four and a half feet of water was maintained above the filter matrix to prevent the formation of air bubbles in the filter matrix during extended periods of filtration. A schematic diagram of the piping arrangements is shown in Figure 11. During filtration, water enters through a float valve at the top which maintains a constant water level. In the backwash mode of operation, water enters the filter at the bottom and exits through a drain trough at the top. Flexibility of operation was provided by a valve and pump through which liquid could be recirculated or transferred to an elevated storage drum. A rotameter (range 0.2-6 gal/min) in the effluent line was used to measure the flow which was controlled by the effluent valve. In the first filter, several drain arrangements were tried, but the simplest and most satisfactory proved to be a short section of rain gutter and a three inch diameter downspout. The filter box was notched to a depth of two inches, as may be seen in the lower left hand section of Figure 12. This arrangement provided more than ample capacity for the available water supply and prevented annoying overflows.

The water supply enters the Dangerous Reactions Laboratory through a three-fourths inch pipe which furnished an adequate volume for backwashing of the 40-60 mesh sand; it was used for the first filter and was sufficient to produce a 100 percent expansion of the sand bed. Even at maximum flow, the support gravel remained undisturbed.

This water supply was inadequate to properly backwash larger-grained bed materials, so the second filter was installed in the Unit

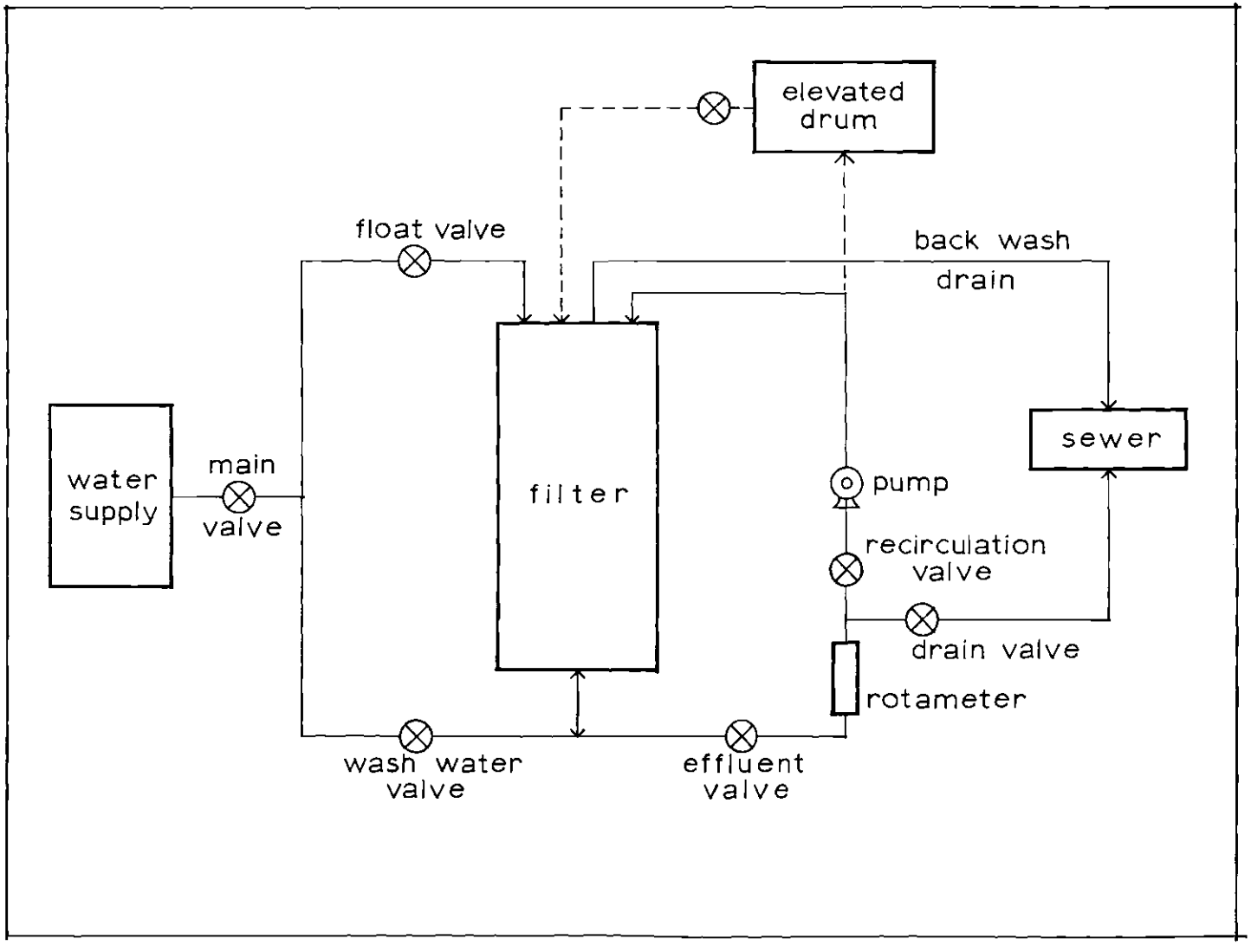

Figure 11. Schematic Diagram of Apparatus

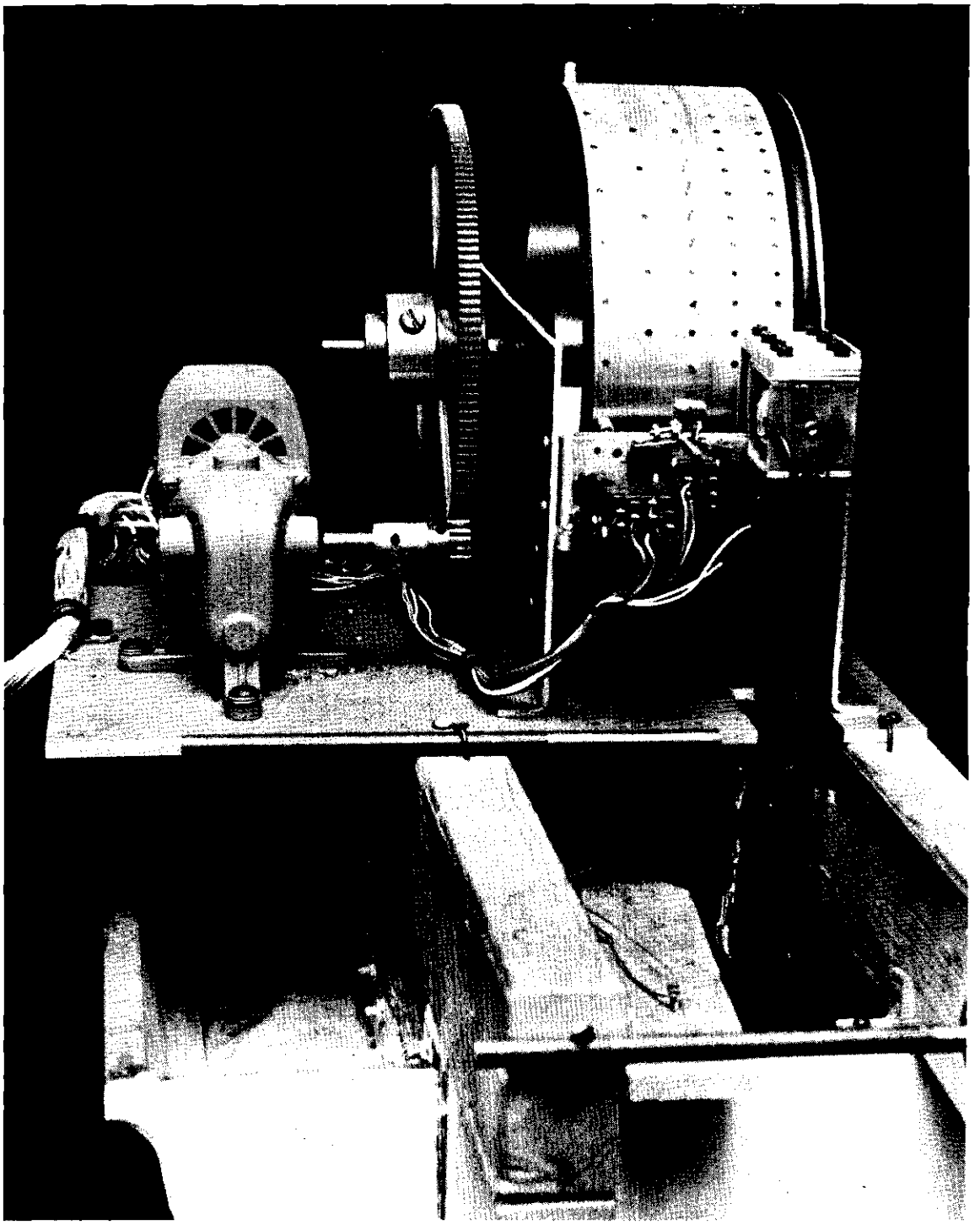

Figure 12. Closeup View of the Programming Drum Mounted on a Filter

Operations Laboratory near a one and one-fourth inch copper line about three feet long, feeding directly from a four and one-half inch cast iron conduit. Even with the head losses brought about by several 90 degree bends and an additional 15 feet of connecting pipe, this supply was quite adequate. Care was necessary during backwash operations to avoid disturbing the bed support material. The flow of wash water through this filter was limited only by the configuration of the drain, and this could have been easily modified if any increased rate of flow had been desired. The drain channel in the concrete floor was coated with melted tar in order to provide a smooth surface and prevent retention of particulates during backwash.

The uniform porous bed was supported on a series of layers of materials decreasing in size from bottom to top. The water opening in the bottom of the box was covered with a coarse screen, and porcelain balls were placed above it. The bottom layer was of porcelain balls about one and three-fourths inch in diameter and was covered with other porcelain balls ranging down to about three-fourths inch in diameter. A layer of one-half inch glass marbles came next, and above this were three layers of crushed quartz pebbles of decreasing size. The height of the support layers totaled about 16 inches. The separation between the bed and the support was quite clean, and even the smallest of the support media was dislodged only under excessive backwash flow conditions.

The electronic system first assembled consisted of a detector plus a manually operated scaler; a second system added a control unit which automatically positioned the detector and operated a scaler. In

both instances the detector was an EON #5114 halogen-quenched cylindrical Geiger tube. The small diameter (five-eighths inch) and short sensitive volume (one inch) of this tube made collimation less difficult than with larger, longer tubes which were first tested. Each detector was placed inside a lead shield containing a horizontal slit. Several shields of different styles were constructed, but the most satisfactory were those of the design shown in Figure 13. These were machined from two pieces of lead which were bolted together with inserted cardboard spacers to regulate the width of the slit. The wall thickness of this style shield is approximately one and one-sixteenth inch, which gives a 96 percent reduction in intensity of the 0.662 MeV cesium-137 gamma. The slit width selected for use was 0.053 inch. This allows the Geiger tube an unshielded view through a solid angle of approximately 4°20'.

In use, the shielded Geiger tube slid inside a vertical rigid transparent plastic tube of suitable inside diameter. The lower end of this plastic tube was plugged and seated in the bed support layers; the upper end extended several inches above the maximum water level and was held in place by a cross brace. The shield and Geiger tube in filter #2 were suspended from the coaxial cable; the weight of the heavier shield used in filter #1 was borne by a wire rope. In each instance a counterweight on the opposite side of a pulley permitted the shielded detector to stay in any position in which it was placed.

Nuclear Chicago Scalers, Model No. 186 and 8775, were used in the manual operation which was maintained in filter #1. These scalers proved to be very reliable during the entire course of the investigation. The sequence of events involved: 1) positioning the Geiger tube, 2) actuat-

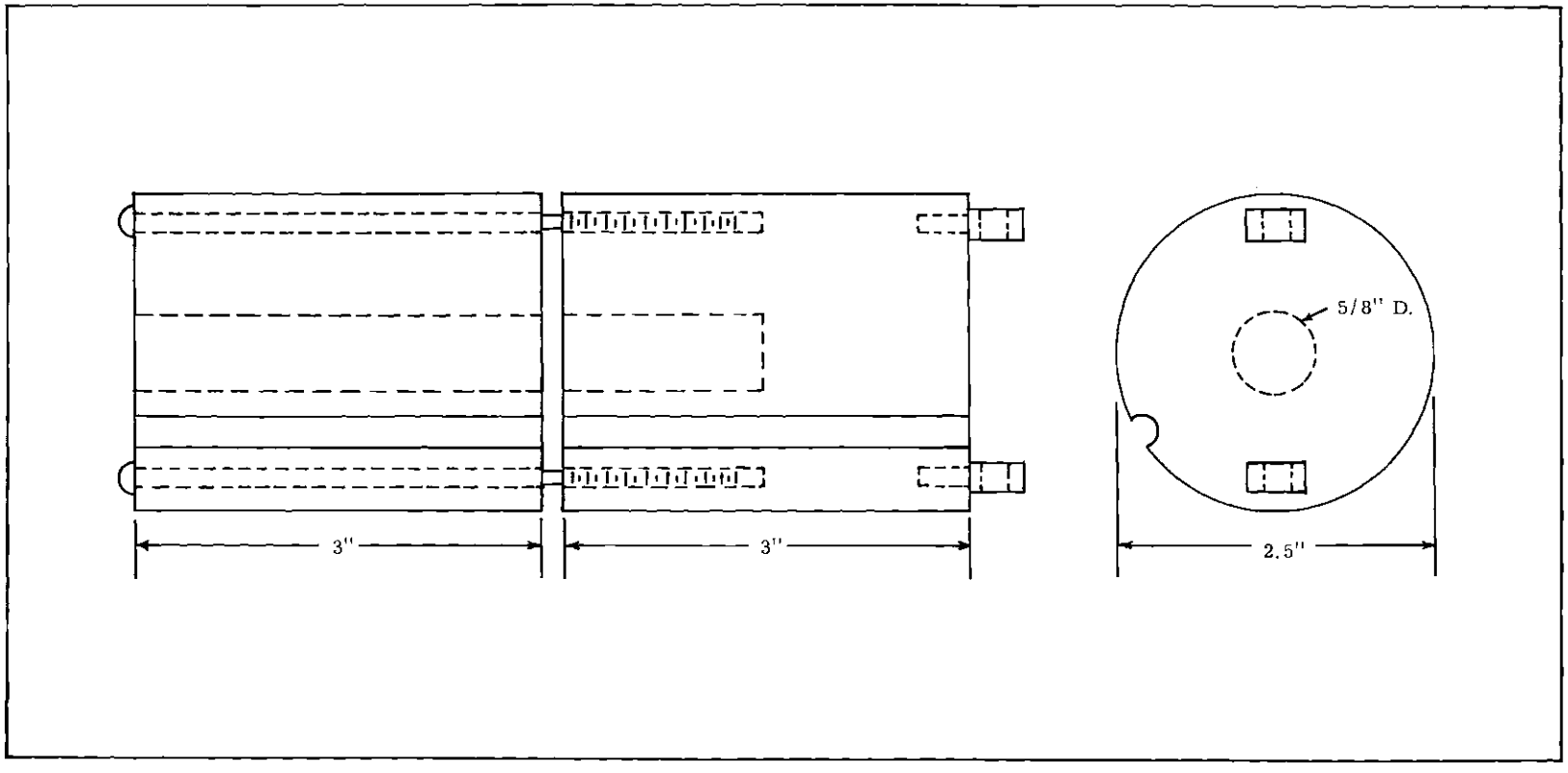

Figure 13. Details of Lead Shield for Geiger Tube

ing the scaler for the selected time interval, and 3) recording the counts measured. This was repeated for each position of interest. Since this routine operation of data taking was found to be both simple and time consuming, it was decided to automate this process as much as possible for filter #2.

An Omni/Guard Low Background Counting System produced by Tracerlab, Inc., Waltham, Massachusetts was available and some of its components were adapted to meet the specific requirements for the automated arrangement. The scaler unit included an anti-coincidence circuit, detector power supply, scaler, and timer. Also utilized was the Tracer/Matic Printing Timer. The coaxial cable which supported the counterbalanced shielded Geiger tube passed over a programming drum, whose position and movement could be precisely controlled (Figure 12). The drum was positively locked in each position by a solenoid-operated plunger so that the selected levels were reproducible. On command, the plunger released the drum which was then rotated by a reversible gear motor to the next desired position. Control of these positions was by microswitches which were actuated by pins in the drum.

When the drum locked into position, a pulse signal started the count determination by the scaler which also furnished the high voltage to the Geiger tube. At the end of the counting interval, the number of counts detected was recorded automatically on the printer. An index number indicating the position of the Geiger tube was also recorded along with the length of the counting interval. After these numerals had been printed, the drum was actuated, the Geiger tube repositioned, and the next level was counted. Following completion of count at all

desired levels, the tube was returned to an initial starting position. A clock-timer was incorporated into the circuit which permitted time intervals between counting sequences in any selected multiple of 15 minutes.

Provision was made for other modes of operation. The timer could be bypassed so that the whole cycle could be begun at any time. In this "manual" mode, the system would continue to recycle until stopped. A switch permitted the system to bypass counting positions as desired, count twice in a given position, or interrupt the sequence and return to the start position. The positioner control unit is shown in Figure 14 and its schematic diagram is Figure 15.

Materials

The suspended matter used in these tests consisted of vermiculite particles prepared from a commercial product supplied through the generosity of The Zonolite Company, Atlanta, Georgia. This commercial material, designated BO-5, is a crude ore mined at Zonolite, S. C. and consists of flakes and fines that have passed through a five mesh screen. The composition of this material is somewhat variable, but a typical chemical analysis is given in Table 4.

An x-ray diffraction analysis was made of the material actually used in this study. It was found that this product is a mixture of regular 1:1 mixed layers of vermiculite and a mica which is known as hydrobiotite, plus irregular mixed layers of hydrobiotite and vermiculite. Identification was based on ASTM x-ray cards 13-233 and 13-465, and it is detailed in Table 5. The x-ray diffraction pattern is shown in Figure 16. This vermiculite ore is very similar to that discussed by

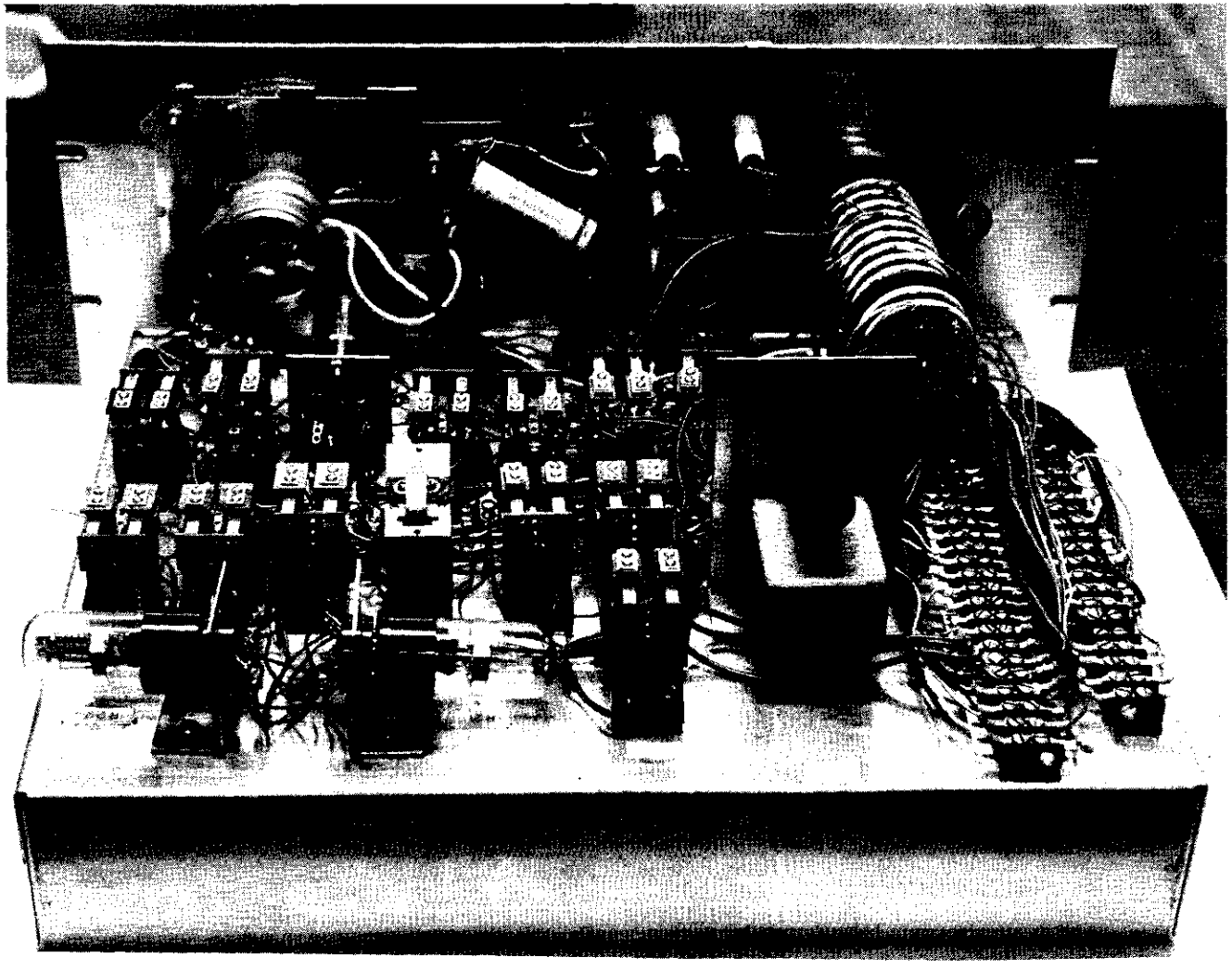

Figure 14. Back View of Positioner Control Unit

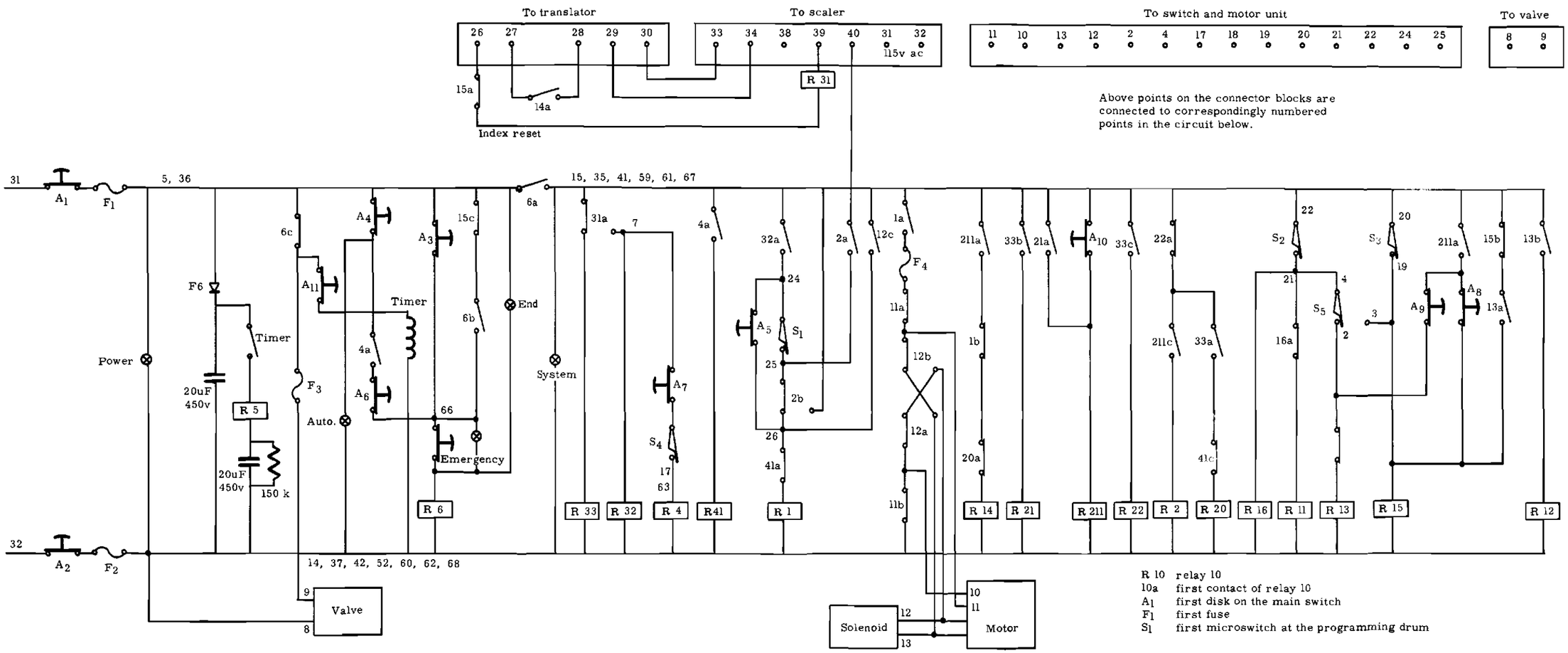

Figure 15. Schematic Diagram of Positioner Control Unit

Table 4. Typical Chemical Analysis of Vermiculite[107]

SiO_2	38.64
MgO	22.68
Al_2O_3	14.94
Fe_2O_3	9.29
K_2O	7.84
CaO	1.23
Cr_2O_3	0.29
Mn_3O_4	0.11
P_2O_5	trace
S	trace
Cl	0.28
H_2O	5.29

Note: Figures shown are percent.

Table 5. X-ray Diffraction Data for Vermiculite Powder

Commercial Vermiculite*		Hydrobiotite†		Hydrobiotite -- Vermiculite‡	
d	I/Io	d	I/Io	d	I/Io
24.5	40	25.5	100	---	---
14.5	90	---	---	15.0	100
12.4	100	12.5	100	12.5	80
8.2	10	8.2	30	---	---
4.92	15	4.92	60	---	---
4.52	5	---	---	4.5	60
3.50	25	3.53	100	---	---
3.36	25	---	---	3.40	40
3.10	25	3.09	100	---	---

*A commercial vermiculite randomly oriented powdered sample with particle size below 25 microns.

†ASTM Card 13-233--"A commercial vermiculite. Flake sample cut from mineral, giving basal reflections only. Regular 1:1 mixed layer vermiculite-mica = hydrobiotite."

‡ASTM Card 13-456--"A commercial vermiculite. Irregular mixed layers of hydrobiotite and vermiculite."

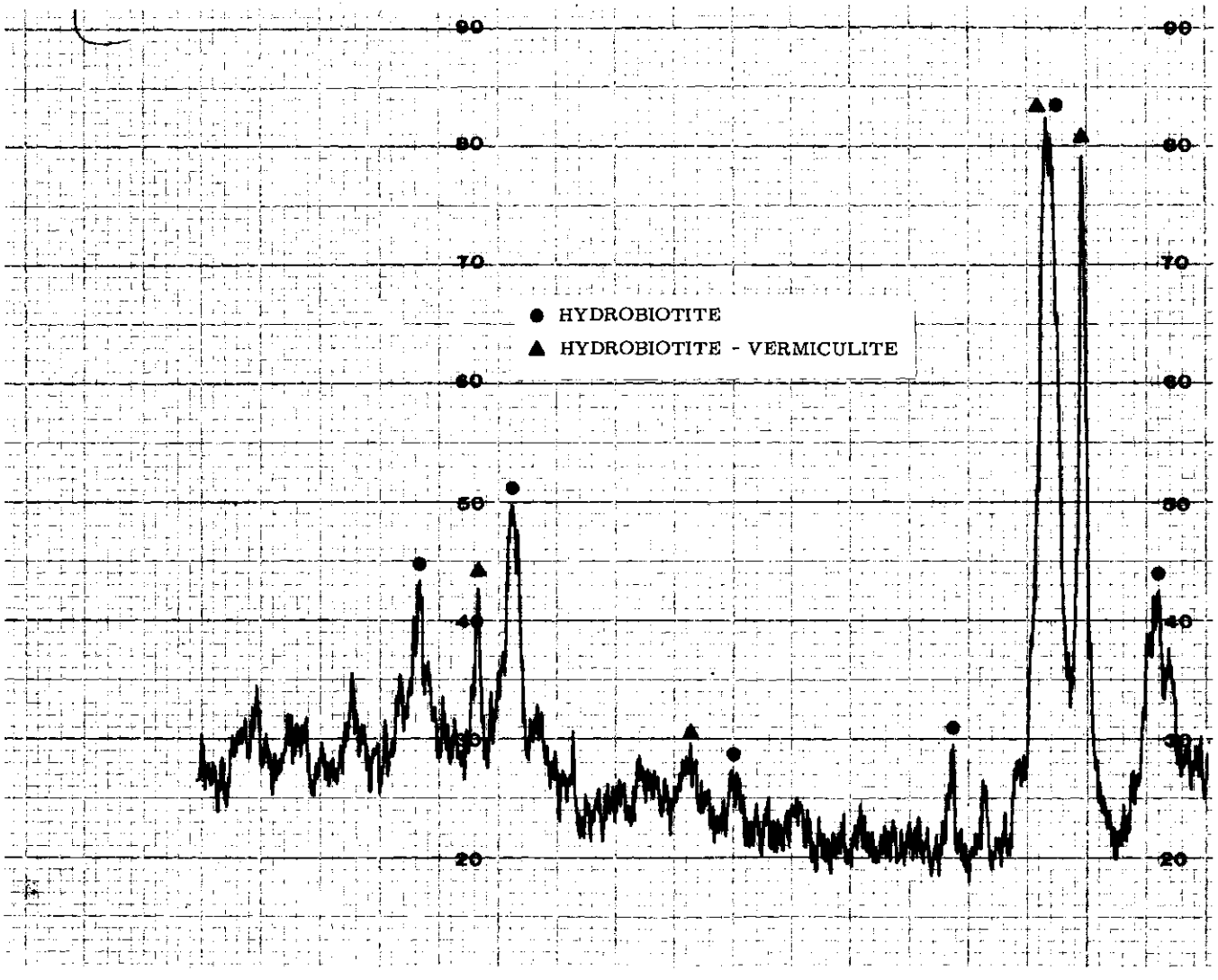

Figure 16. X-ray Diffraction Pattern of Ground Vermiculite

Midgley and Midgley.[107]

After an initial dry screening of the material as received, the portion retained on a U. S. Standard Sieve No. 170 (openings 88 μ) was reduced by ball milling. Excessive milling rapidly produced a high percentage of particles so small that they quite easily passed through the finest screen used, so it was necessary to mill for a number of short intervals and sieve after each milling interval. The ground material was screened through a series of standard sieves having openings of 88, 74, 63, 53, 44, 37, and 25 μ.

The sieves were agitated by a vibrator but prolonged periods of vibration were necessary to achieve passage of appreciable amounts through the finer screens. Examination under the microscope of dry screened fractions revealed the presence of much material both larger and smaller than the nominal fraction size. Wet screening under running water removed much of the finer particles. Size distributions were determined by measuring particle diameters under the microscope. Figure 17 shows a typical distribution for material at this stage of preparation. As discussed below, activity is proportional to volume, so the relative effect of the numerous small particles is not very great. This is also shown in Figure 17 by the curve representing relative activity. It is possible to eliminate most of these small particles by swirling the batch in water, allowing it to stand a few seconds, and then decanting. Some of the larger granules are also carried away in the decantate, but the size distribution of the remainder is quite narrow. It should be noted that a similar situation also exists with respect to larger mesh screens, and screened granular materials typically are larger than nominal screen

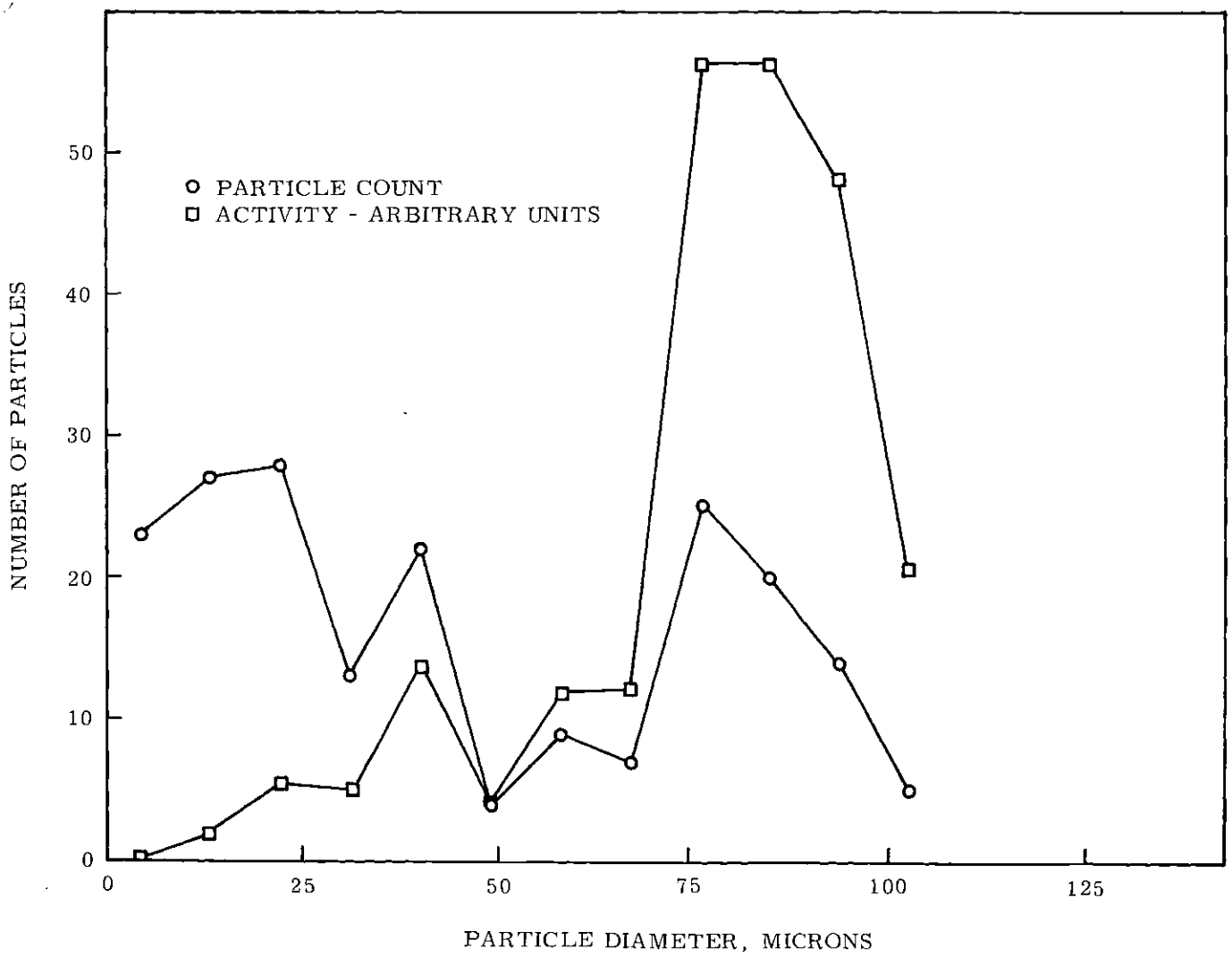

Figure 17. Particle Size and Activity Distribution of 44-53 Micron Vermiculite

mesh designations.

The sand used initially in these experiments was supplied by the Atlanta Water Works. The exact history of this sand is not known, but it was first installed in a rapid sand filter at the Hemphill Treatment Plant in 1926. It continued in use until either 1941 or 1948, at which time it was removed to make room for surface wash equipment. Since removal, it had been stored on the ground beneath the sand filters. A sieve analysis of this sand was made and the resulting curve is shown in Figure 18. The 20-30 mesh fraction of this sand was separated by hand screening and was used in the Series I experiments.

The Pennsylvania Glass Sand Corporation, Atlanta, Georgia graciously supplied two different size ranges of clean white silica sand from their plant at Columbia, S. C. A typical chemical analysis of this sand is reported in Table 6, and screen analyses of both sizes (designated #1 and #3) are shown in Figures 19 and 20, respectively. The #1 product was screened to obtain 40-60 mesh material, and the #3 grade was sieved for the 14-20 mesh fraction.

The ground anthracite used was supplied by the Palmer Filter Equipment Company, Erie, Pennsylvania. This product, which is tradenamed "Anthrafilt," is prepared from a selected Pennsylvania anthracite which has been cleaned so as to have a minimum ash content. It is then screened and classified hydraulically to yield sizes suitable for filter purposes. This product is available in a number of different grain sizes, but the material used here is designated No. 1 and was reported to have an effective size in the range 0.60-0.80 mm and a uniformity coefficient of 1.85. The "effective size" is the particle size in the

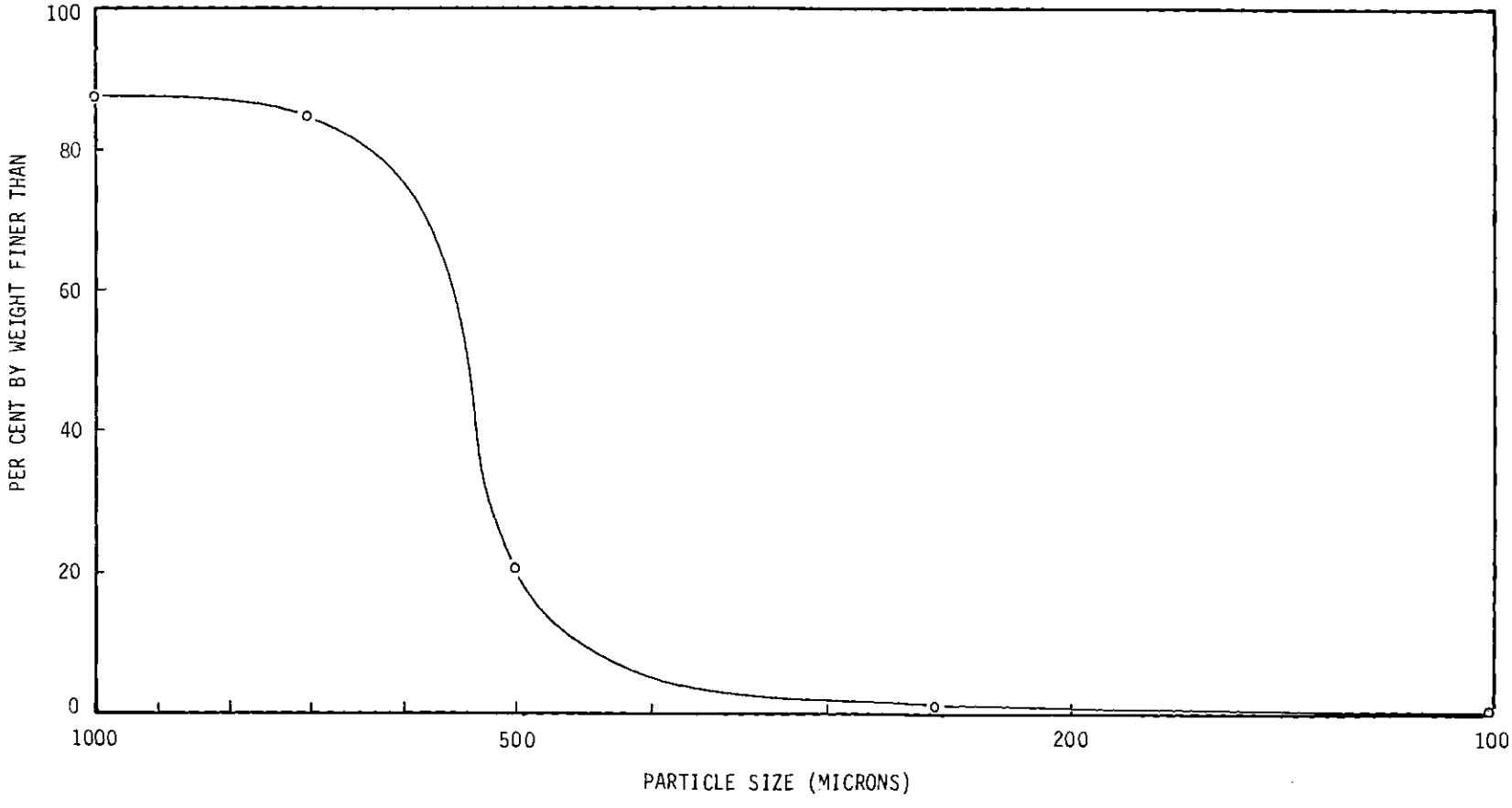

Figure 18. Particle Size Distribution of Atlanta Water Works Sand

Table 6. Typical Chemical Analysis of Sand #1 and Sand #3 Supplied by Pennsylvania Glass Sand Company

SiO_2	99.77
Fe_2O_3	0.014
Al_2O_3	0.152
TiO_2	0.018
CaO	trace
MgO	trace
Loss on ignition	0.08

Note: Figures shown are percent.

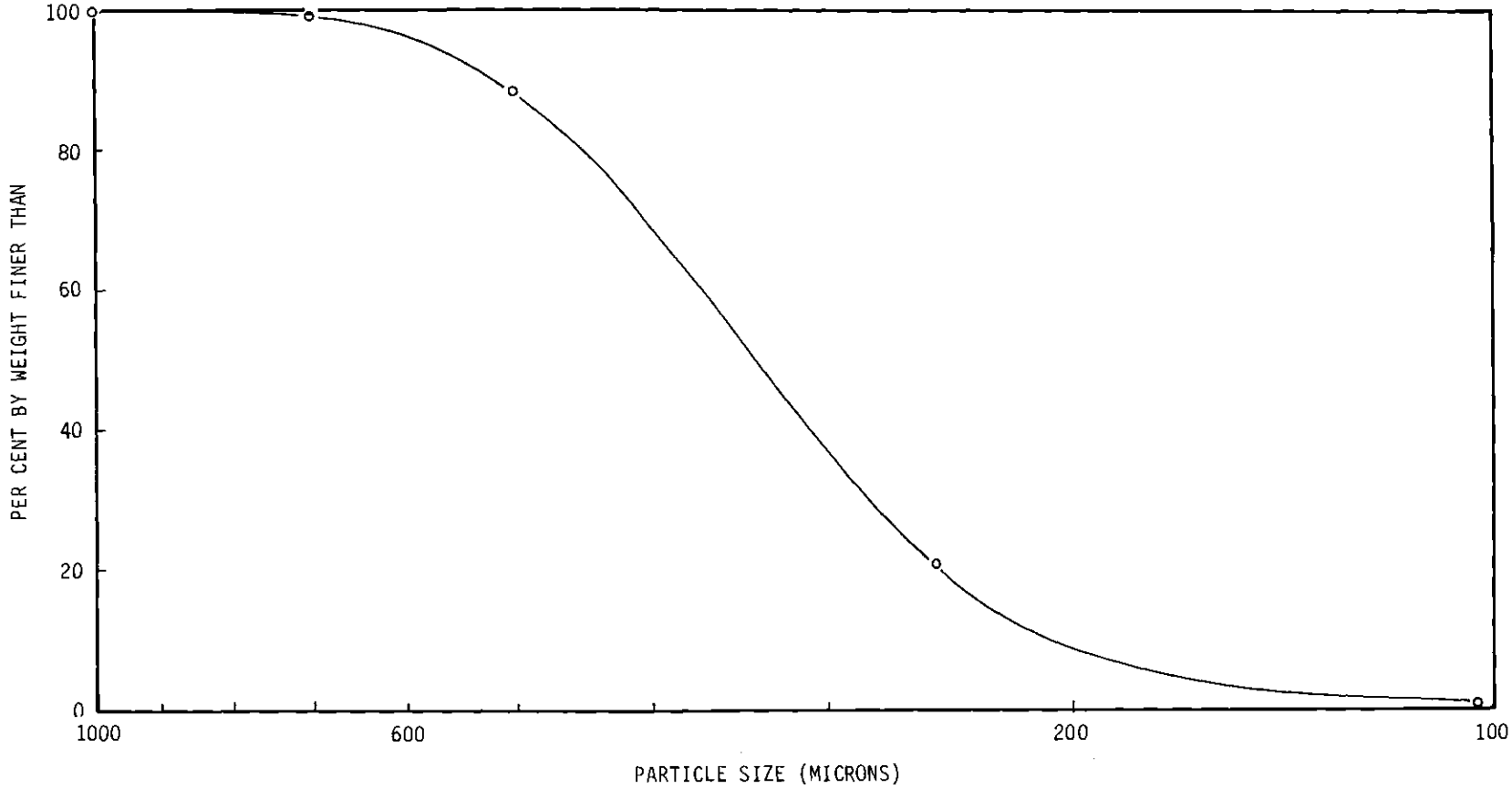

Figure 19. Particle Size Distribution of Pennsylvania Glass Sand Corporation Sand #1

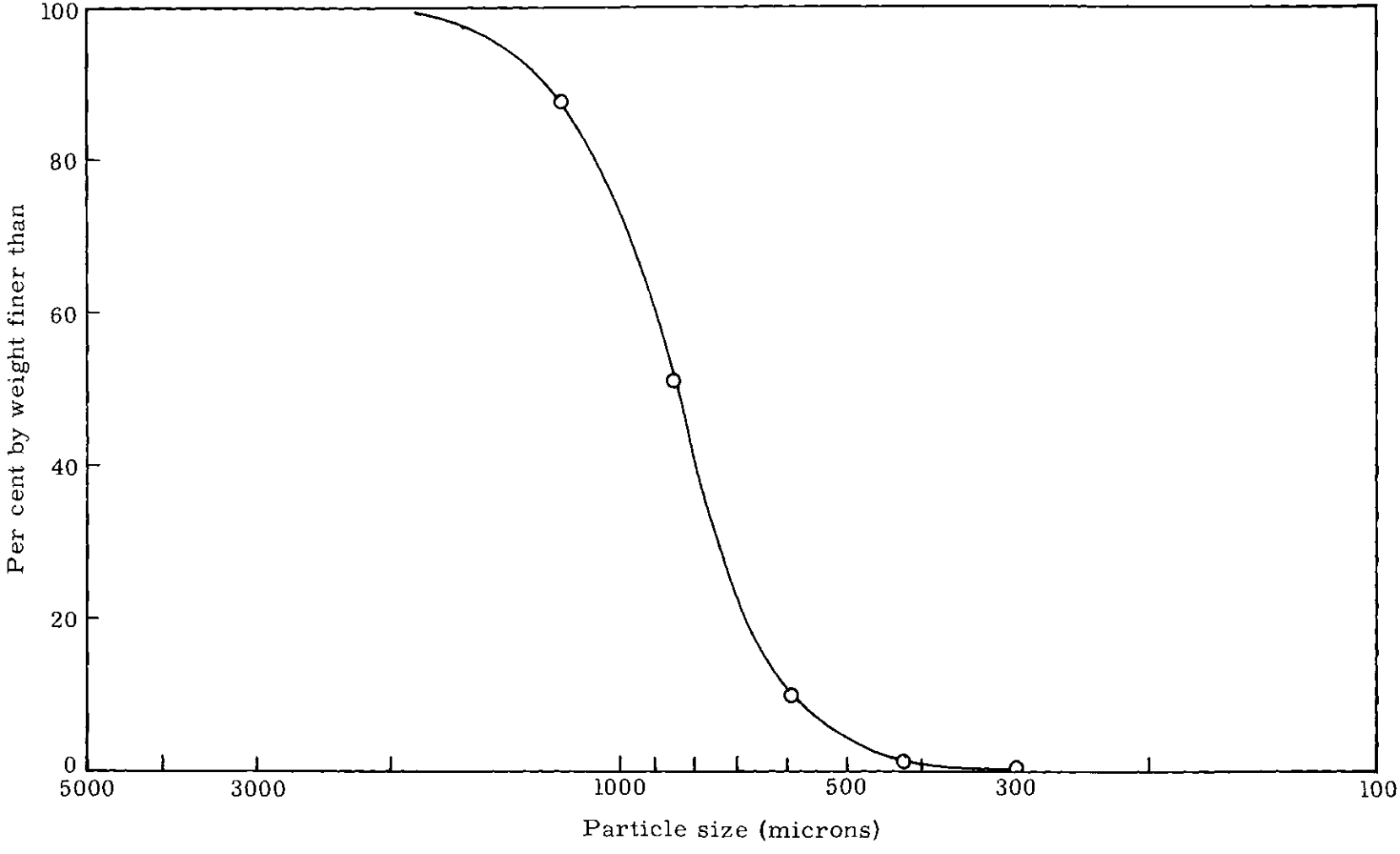

Figure 20. Particle Size Distribution of Pennsylvania Glass Sand Corporation Sand #3

distribution which is larger than 10 percent of the total. The uniformity coefficient is the 60 percentile (60 percent smaller than) divided by the 10 percentile (10 percent smaller than).

A screen analysis of the Anthrafilt actually used revealed the distribution shown in Figure 21. From this, the effective size was found to be about 0.65 mm, and the uniformity coefficient 1.85 in excellent agreement with the specifications.

Radioactive Labeling Procedure

Vermiculite has a relatively high cation exchange capacity (> 1 meq/g) which facilitates labeling with a radioactive cation. Cesium-137 was chosen as the labeling isotope because of its long half life, ease of detection, and ready availability. The procedure consisted of stirring the screened particles in a solution of cesium-137 chloride.

By experiment, it was found that approximately 20 μCi of cesium-137 in the filter at any one time gave a satisfactory count rate, and this much activity could be conveniently carried by one-half to one gram of vermiculite. Each complete series of particle sizes was prepared from a five gram batch of ground vermiculite from which the fraction smaller than 25 μ diameter was removed by screening. The batch was dispersed in about 300 ml of water to which 150 μCi of Cs^{137} had been added. Stirring was continued for several hours, and the mixture allowed to stand overnight or longer. The particles acquired the radioactive label essentially by an ion exchange process and were introduced into the filter system after being washed briefly to remove non-adhering cesium tracer. In the course of several tracer experiments, it was observed

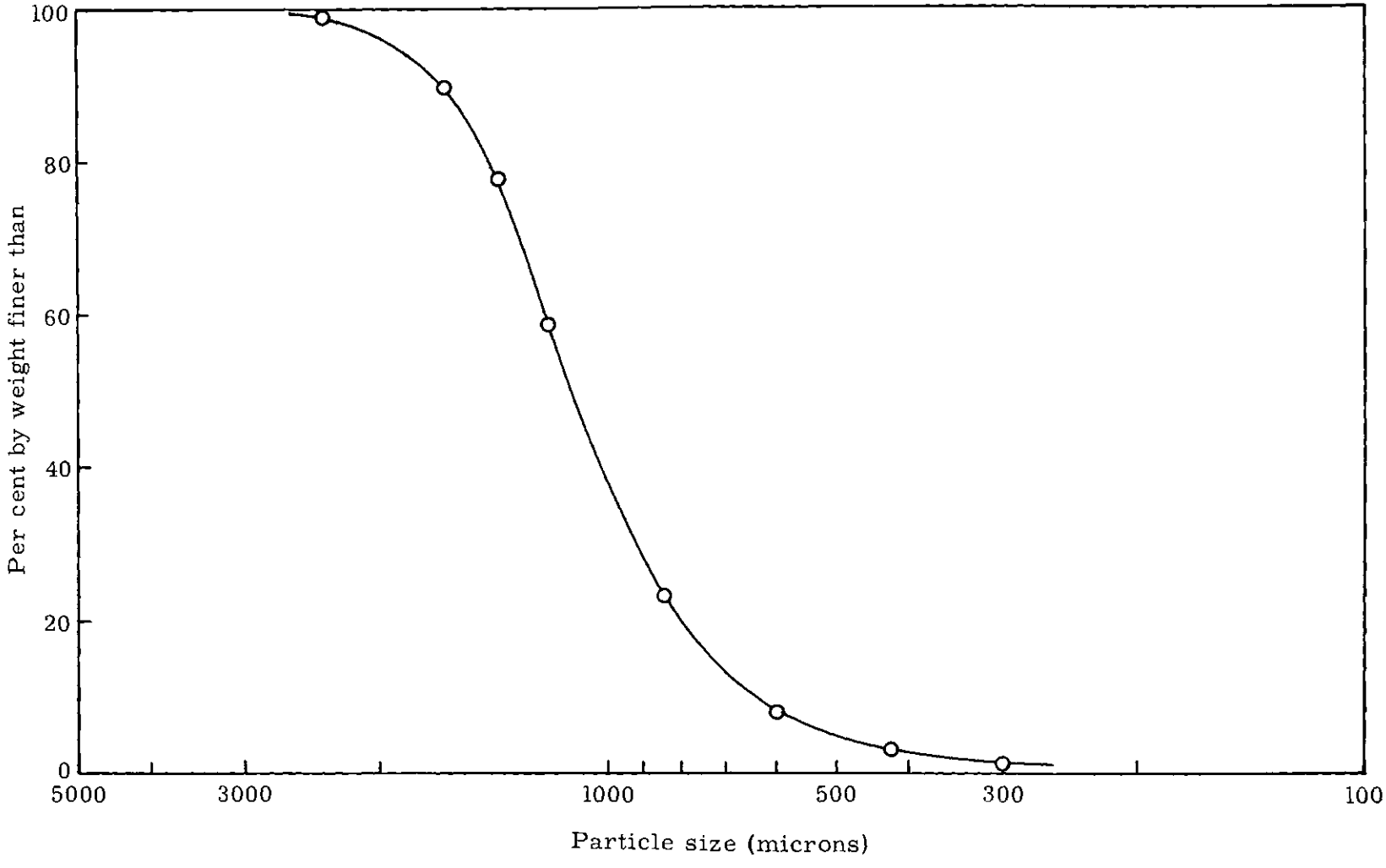

Figure 21. Particle Size Distribution of Anthrafilt No. 1

that, after reaching a maximum, the total activity in the filter decreased due either to passage of the labeled particles through the filter or due to loss of some of the radioisotope tracer from the particles. It was found experimentally that the second alternative was definitely occurring, although the filter effluent never revealed a detectable amount of dissolved activity when checked continuously with a Geiger counter or when batch samples were placed in a well-type scintillation detector. Appreciable activity was accumulated, however, by passing the effluent through a layer of cation exchange resin.

Analysis of activity profiles in the filter bed would be simplified by eliminating this variable, so calcining of the vermiculite particles was investigated as a means of fixing the tracer. The sized vermiculite particles were labeled in the usual fashion by stirring in Cs^{137} solution. After decanting most of the solution and washing a few times with water, the remaining moisture was evaporated under a heat lamp, and the dry, labeled vermiculite particles were heated in a small muffle furnace. It was quickly determined that heating to an indicated temperature of less than 1850° F had no effect on the tenacity with which the isotope was held, and that temperatures in excess of about 2300° F fused the particles into a solid single mass. Between these limiting temperatures, it was possible, however, to obtain a product only lightly fused and that could be broken apart with a spatula and by grinding briefly in a mortar. It was not found possible to obtain calcined -25 μ particles directly. At temperatures high enough to fix the activity (about 2050° F), the -25 μ particles fused solidly; this small size particle was obtained by grinding larger calcined material in a mortar.

Particles with a diameter of one to two microns were prepared by ball milling for 20 hours. The grinding process does not affect the concentration of activity of the particles, as the cesium-137 is apparently held throughout the carrier, not simply on the surface. This was shown by the results of measuring activity of the screened, dried particles. It was found that activity was proportional to the total weight of vermiculite, regardless of particle size.

After calcining and trituration, the particles were rescreened under running water to insure uniform size fractions. Preliminary runs were made using calcined particles. During the first few minutes of flow, the activity increased to a maximum in typical fashion, but thereafter remained essentially constant. Filtration was continued in one instance for more than 10 days without diminution of the activity level. Subsequent runs have confirmed that calcination attaches the cesium-137 activity quite satisfactorily to vermiculite particles and even prolonged rapid water flow does not elute the activity.

Test Procedure

Before each run, the filter was backwashed by admitting water to the bottom of the filter box which caused the bed to expand upward and become fluidized. As the flow pattern became stabilized, the upper limit of bed material remained at a constant level which depended on the flow rate.

Most efficient washing of sand is reported[48] to occur at a bed expansion of about 20 percent. Under such conditions, the sand grains are in rapid random motion and particulate matter trapped by the filter

is flushed out. The cleansing of the sand is greatly facilitated by the contact of the grains with each other. This is apparently a mechanical action and simply involves knocking or scraping off any adhering particles. The amount of water used for backwash is an important variable in commercial water filtration, but it was of little concern in this study; backwash periods of 20 to 30 minutes were normally used.

At the conclusion of backwash, the sand settles into a metastable configuration of greater height than will exist after filtration has proceeded for a time. Therefore, in order to have a fixed, reproducible compaction of sand, the regular procedure involved striking the outside of the filter box with a rubber mallet as the sand was settling. This rapidly produced a more stable configuration that did not change even during prolonged filtration runs. The sand was returned each time to the same elevation, so that the degree of compaction was considered constant during the entire period of operation.

After backwash, downward water flow in the filtration mode was started and a determination of the background radioactivity was made. This was accomplished by placing the detector at each level in the filter to be measured in turn and counting for a selected interval. The counting interval was usually 10 minutes, but an occasional series of 30 minutes or more was run. In general, the background level of radioactivity remained about the same, but it was subject to a gradual increase when uncalcined or insufficiently calcined labeled particles were used. If the background reached an undesirably high level, it was reduced by rinsing the bed with a dilute hydrochloric acid, followed by a thorough backwash.

The selected fraction of previously calcined and screened vermiculite particles (which were stored under water) was thoroughly dispersed by placing them in a small beaker in an ultrasonic bath for a short time. The particle suspension was then poured into the top of the filter where water flow was continuing in the filtration mode. Experience showed that activity at the sand surface or in the sand continued to increase until all the radiotracer was removed from the water being filtered. Some mixing occurs in the water volume above the porous bed, and about 20 to 30 minutes was required for the activity to reach a maximum.

Measurement of activity at the bed surface and at each selected elevation within the bed was then made. This was accomplished by placing the Geiger detector at the appropriate location and counting for a selected interval, usually 10 minutes. This was done either by hand or by the automatic positioner-counter control unit. Measurements at the same levels were subsequently repeated many times to determine if there was any change in the indicated pattern of deposition and to accumulate statistically reliable data. At no time was there any indication of change of the deposition pattern within the filter matrix.

The background count at each level was subtracted from the average observed count corresponding to that level. The remaining net count was taken as proportional to the amount of activity, and therefore to the quantity of tracer particles in the bed at that elevation.

After completion of a run, the filter was backwashed, allowing the labeled particles to be washed down the drain. They were accompanied and followed by a very large quantity of water, and the resulting waste

stream had a maximum activity below the level of detection by a Geiger-type survey instrument. The calculated activity in the waste was well within the minimum permissible concentration, and it is believed that no hazard was created by this means of disposal.

CHAPTER IV

EXPERIMENTAL RESULTS

Counting Geometry in the Filter

As a preliminary prerequisite to interpretation of the data accumulated by counting at each level of the filter, it was necessary to ascertain the relationship between activity and, therefore, particle concentration, at each level and the observed count rate. This was determined in a positive manner by removing sand samples from the filter at the conclusion of a run and analyzing them individually. It was necessary to first drain the filter, and it was assumed that drainage caused no significant displacement of the deposited particles. This assumption was validated by a series of readings taken before and after draining. The results are shown in Table 7. All readings taken at the same level after draining are higher than prior to draining, due presumably to the reduction in attenuation by the water as compared to the air which replaced it. The ratio of values between levels in each case is about the same.

The plastic window above the sand bed was taken off the filter after draining, and sand samples were removed with a piece of stainless steel sheet, bent to form a channel. The rectangular core recovered with this tool could be easily sliced into sections of desired thickness. The individual slices were separately dried, homogenized, and counted with a scintillation detector and scaler. The type of curve

Table 7. Gross 10 Minute Counts Observed in 14-20 Mesh Sand

Sand Depth (inches)	Before Draining	After Draining
0	1306	1402
1	1029	1200
2	619	664
3	375	488
4	224	276
5	139	183
7	103	118
9	99	140

resulting from a plot of counts versus increment position is shown in Figure 22. Superimposed is a line illustrating the corresponding Geiger tube readings taken in the filter prior to removal of the sand samples. While the fine details of the actual curves are interesting, they are obviously greatly dependent on the thickness of the individual increments taken for measurement. The ratio of in situ counts to those found in the sand taken from the same level is not constant for the elevations examined. However, the number of counts detected in each sand sample decreased with increasing depth of the filter; counts observed in situ also decreased. As only relative values are of concern here, this correlation is adequate.

One of the problems peculiar to the present experiment is the fact that the activity in the surface layer may greatly exceed that in the bed volume below. As a result, the count rate observed in layers only a few inches below the surface may be greatly affected by the attenuated but still significant contribution of the surface layer activity, which may vary from run to run.

To establish the importance of this effect, an extensive series of calibration experiments was performed to correlate deposited activity with the number of counts detected. It was also important to determine the solid angle of bed volume seen by the detector. For this purpose, a plastic bucket containing two parallel vertical tubes was filled with sand as shown in Figure 23. Inside one tube the shielded and collimated Geiger tube could be raised and lowered. Inside the second tube, which was spaced two and seven-eights inch center to center from the first, small sources of radioactivity of known relative concentration were

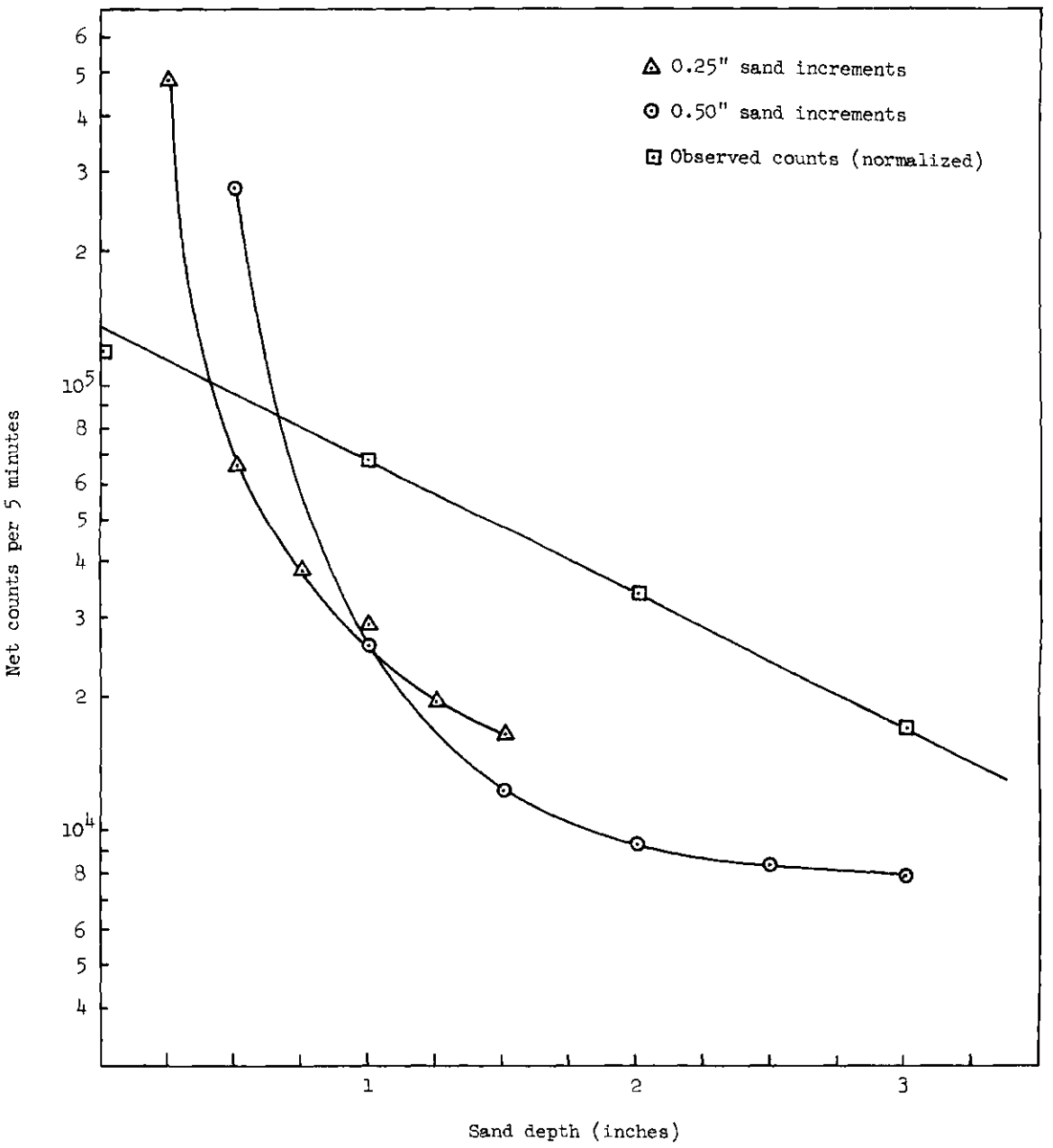

Figure 22. Count Variation with Depth of Samples Removed and *in situ*

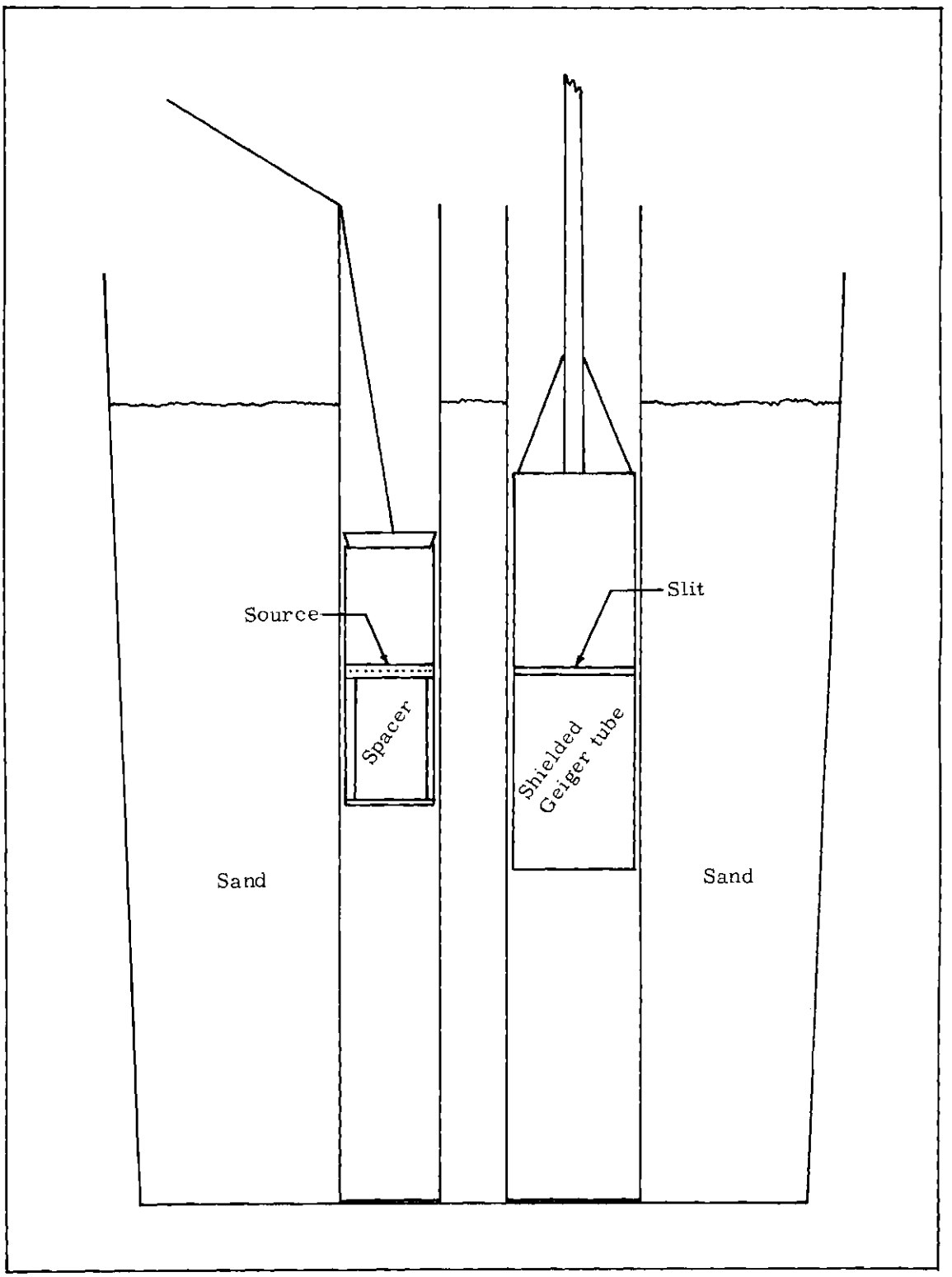

Figure 23. Diagram of Sand Bucket Experimental Arrangement

placed. These radioactive sources were prepared by evaporating a small amount of cesium-137 solution in a planchet. A slightly smaller planchet was then pressed into the first, neatly sealing the activity between the two. A number of these sources with strengths ranging over two orders of magnitude were prepared. Their relative strengths were determined with a scintillation crystal detector and scaler.

In use, these sources were arranged vertically with inserted spacers so that the vertical separation distance was one or more inches. This simulated the pattern of activity distribution in the filter, except the contribution of each layer was concentrated into a plane rather than being spread over an appreciable depth. The volume of observation of the detector was easily determined by placing a single radioactive source at a depth of three or four inches below the sand surface and observing the count rate at various elevations as the detector was lowered incrementally down its guide tube. The near symmetry of the observation field is shown in Figure 24, which was prepared from data gathered in this manner.

A series of measurements was made with two sources spaced vertically one inch apart. To simulate as closely as possible the conditions existing in the filter, the upper source was aligned with the surface of the sand and counts observed at one inch intervals beginning at the surface. Various pairs of sources were used so that the curves resulting from different ratios of strength could be compared. A summary plot of most of these results is given in Figure 25. The number associated with each curve is the ratio of strength of the source at the sand surface to the strength of the source at the one inch depth in the sand.

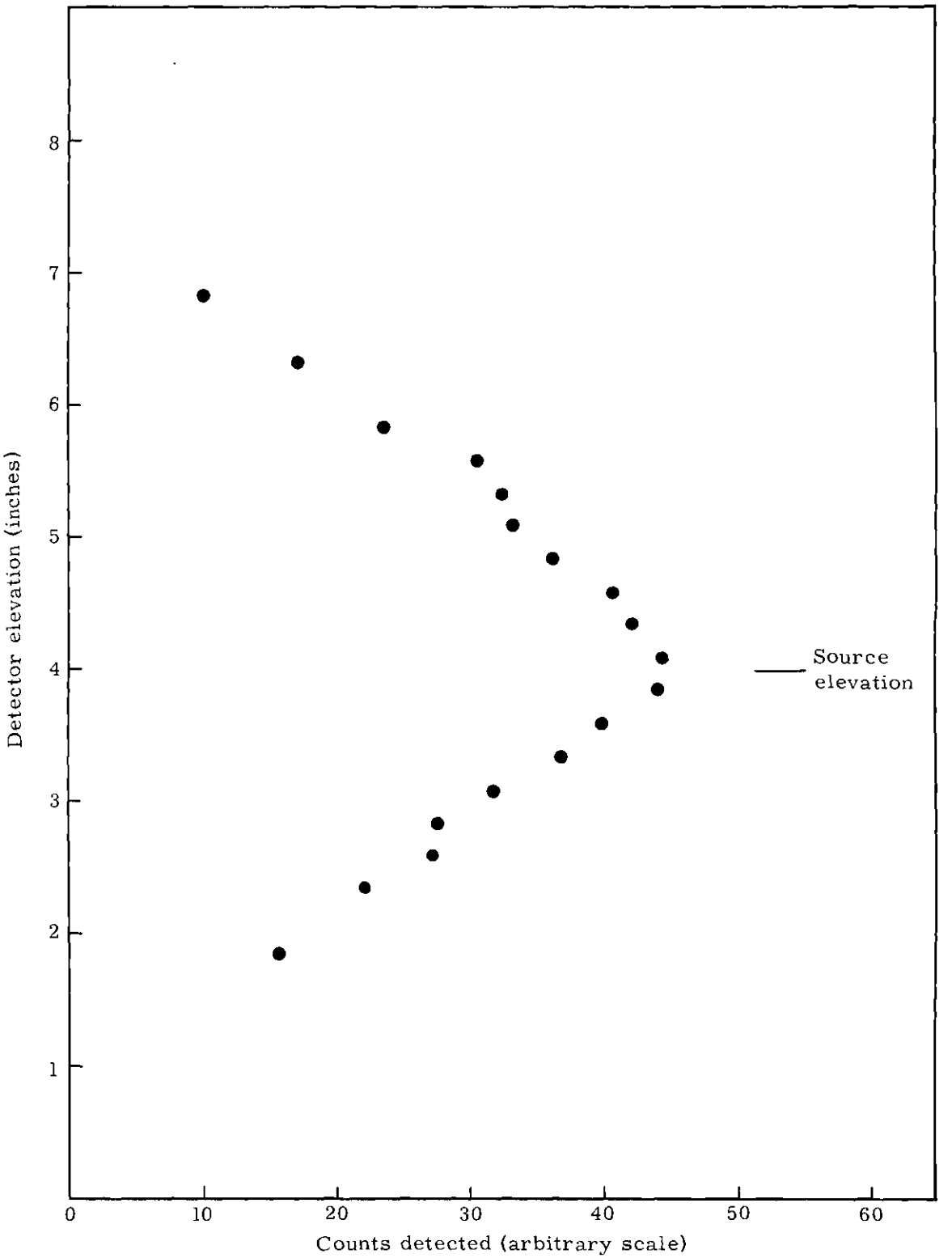

Figure 24. Field of Observation of Shielded Collimated Geiger Tube

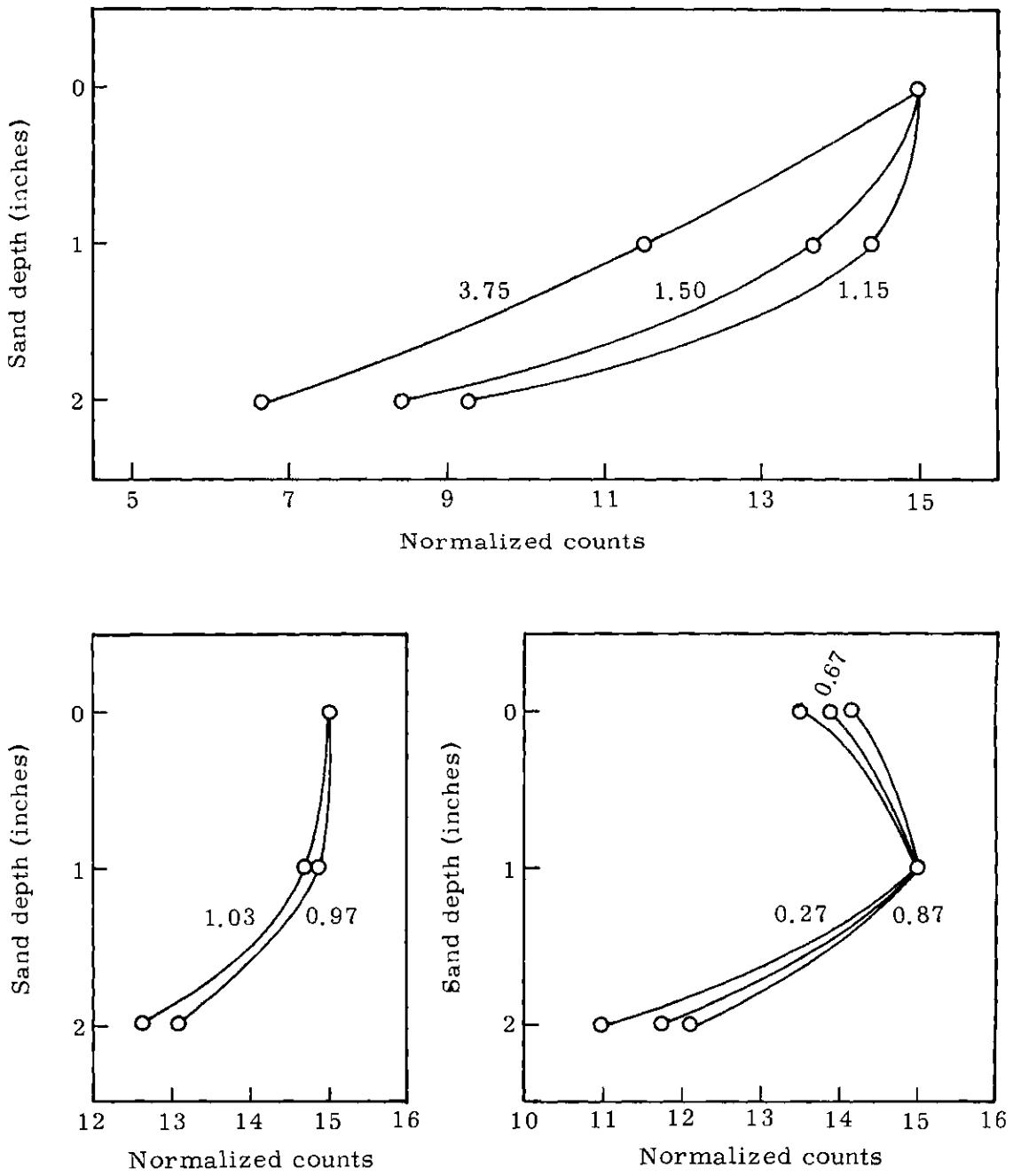

Figure 25. Activity Profiles for Various Source Strength Ratios

It can be seen from Figure 25 that the shape of the curves is generally rather insensitive to the ratio of activity between the surface and the one inch depth, and it is only in those instances where the sources differed greatly in activity that the curves vary greatly.

The shape of these curves is due to a combination of at least three factors: the geometry of the collimating shield and slit, square-law fall-off and attenuation, and degradation of energy due to increased scattering at greater distances through the intervening sand. Figure 26 was prepared from the results obtained by substituting a scintillation crystal for the Geiger tube in the bucket of sand and measuring the resulting spectrum at several elevations with a pulse-height analyzer. These curves were visually normalized by counting until the height of the 0.662 Mev peak reached the same selected point at each counting position.

The source and detector were placed at the same elevation for the initial spectrum, and the detector was then repositioned at different levels for subsequent measurements. The amount of sand between the source and the detector was thereby increased for each subsequent point of observation, and the intensity of detected low energies increased relative to the 0.662 Mev peak. Figure 26 thus illustrates the increased Compton scattering as the thickness of intervening sand is increased.

From consideration of the various factors involved, it is apparent that two localized sources of activity cannot very accurately represent a volume containing activity that varies in concentration with its elevation. Nevertheless, the shape of the curves obtained is informative and aids in the interpretation of results obtained from actual filter runs.

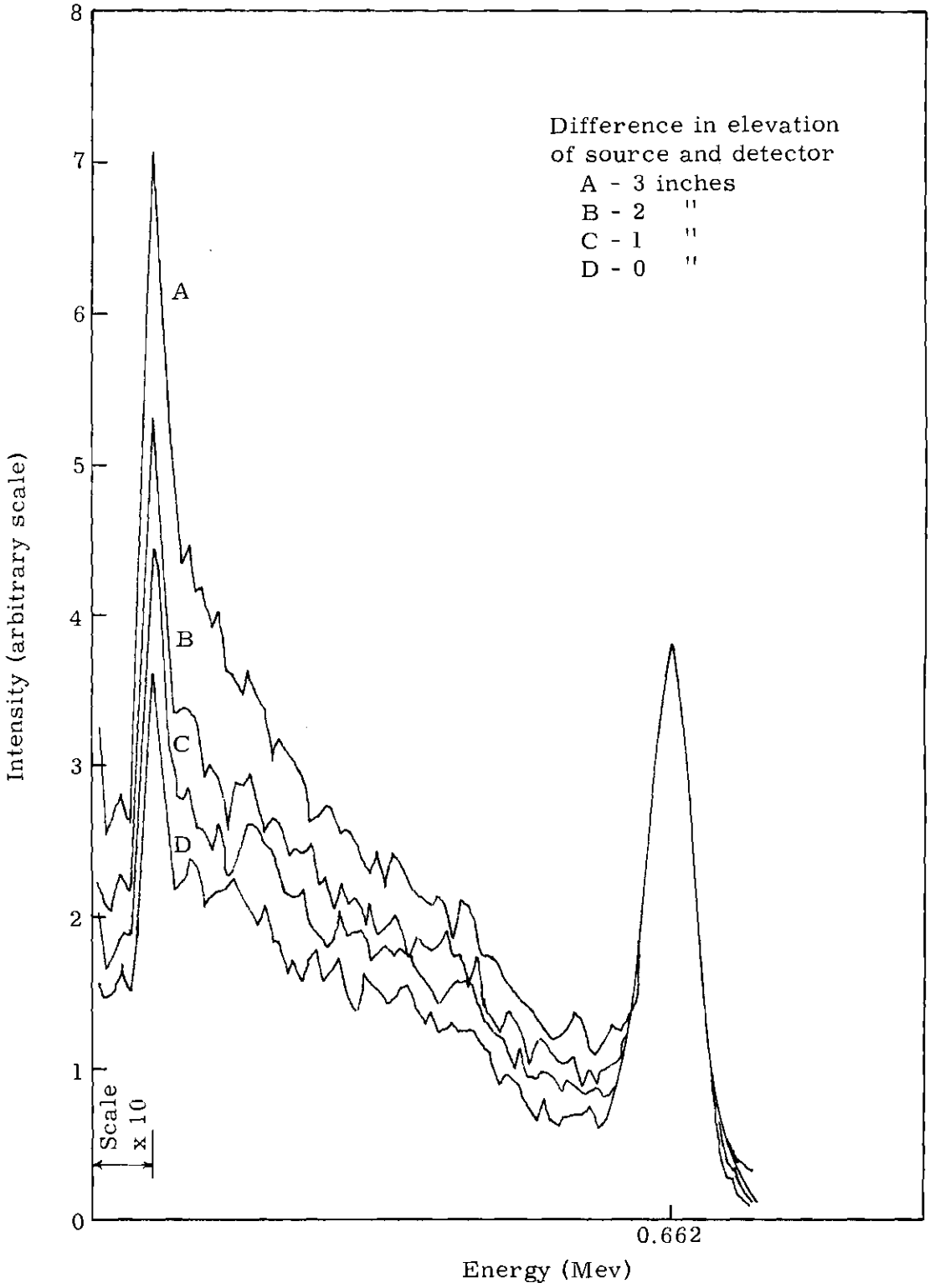

Figure 26. Cs^{137} Spectra at Different Spacings

Determination of Filter Coefficients

The filter coefficient, λ, was calculated from the equation $C = C_o e^{-\lambda L}$, where C_o is the concentration of particles at the surface of the bed ($L = 0$) and C is the concentration at any depth L within the bed. Values of λ were calculated from the relative number of counts observed at the surface and at a depth of one inch. These two points were used because in every case they revealed the highest count rates, giving numbers statistically more reliable. Also, the difference in λ due to different particle sizes is most pronounced when λ is calculated for the top inch of filter bed.

Typical net counts at the surface were in the range of 1000 to 2500 per 10 minutes. For convenient comparison, the results of each run were all normalized to a uniform 1200 at the surface level. The normalization factor for each run was obtained by dividing 1200 by the net counts observed at the surface. Each observed value in the run was then multiplied by this factor.

Referring to the summary of normalized data in Table 8, it will be seen that there is less fluctuation in the values for the one inch depth than for other depths measured. The trend of the data at other levels is in general accord with that at the one inch depth though a few values appear out of place.

Table 9 summarizes the λ values calculated from the data of Table 8. A number of runs were repeated in order to ascertain reproducibility of results and to verify an occasional seemingly anomalous result. In general, such doubtful results could be traced to failure of the automatic positioning drum to lock at the desired bed elevation.

Table 8. Summary of Observed Data*

Bed Depth (in.)	Particle Size (μ)									
	1-3	-25	25-37	37-44	44-53	53-63	63-74	74-88	88-105	105 +
Series I: 20-30 Mesh Sand										
0	1200	1200	1200	1200	1200	1200	1200	1200	1200	1200
1	1187	879	743	705	682	662	640	611	596	587
2	886	462	348	340	355	299	280	268	245	255
3	648	218	143	145	175	130	125	122	106	85
4	488	103	74	75	84	54	52	45	50	63
5	380	51	21	23	39	33	22	39	20	14
7	252	---	---	---	---	6	---	---	---	16
9	182	---	---	---	---	0	---	---	7	11
Series II: 40-60 Mesh Sand										
0	1200	1200	1200	1200	1200	1200	1200	1200		
1	1128	692	583	607	584	587	635	552		
2	740	340	235	193	241	256	283	238		
3	441	170	73	104	101	113	104	108		
4	270	104	29	73	57	106	57	41		
5	159	64	---	42	30	61	29	20		
7	70	24	---	35	16	30	14	17		
9	18	26	---	39	12	57	30	---		

*Counts per 10 minutes normalized to 1200 at the bed surface.

Table 8. Summary of Observed Data (Concluded)

Bed Depth (in.)	Particle Size (μ)								
	1-3	-25	25-37	37-44	44-53	53-63	63-74	74-88	88-105
Series III: 14-20 Mesh Sand									
0	1200	1200	1200	1200	1200	1200	1200	1200	1200
1	1612	1260	949	966	789	737	676	656	674
2	1532	950	506	552	365	514	285	283	298
3	1318	597	244	256	165	247	140	133	125
4	1134	369	120	128	77	205	84	59	62
5	920	222	63	58	40	161	60	28	46
7	659	93	29	12	10	156	44	16	23
9	509	62	23	9	8	166	42	17	13
Series IV: Anthrafilt No. 1									
0	1200	1200	1200	1200	1200	1200	1200	1200	1200
1	1451	984	877	827	774	752	733	726	732
2	1198	586	467	440	376	356	320	332	336
3	923	309	225	207	181	161	164	197	161
4	714	164	109	121	92	60	64	67	71
5	589	86	48	59	47	45	27	70	20
7	368	---	---	12	11	9	---	---	4
9	291	2	5	13	3	---	13	---	---

Table 9. λ Values at 2 gal/ft^2/min

Particle Size	40-60 Mesh Sand	20-30 Mesh Sand	14-20 Mesh Sand	Anthrafilt No. 1
1-3	0.06	0.01	-0.31	-0.19
0-25	0.55	0.31	-0.05	0.20
25-37	0.72	0.48	0.23	0.31
37-44	0.68	0.53	0.30	0.37
44-53	0.72	0.56	0.42	0.44
53-63	0.71	0.60	0.49	0.47
63-74	0.64	0.63	0.57	0.49
74-88	0.78	0.68	0.60	0.50
88-105	--	0.72	0.57	0.49
> 105	--	0.71	--	--

The statistics of the results were investigated through calculation of deviations in λ due to fluctuations in the observed number of counts at each point. The standard deviation of $e^{-\lambda}$ was calculated from the equation

$$\sigma_{e^{-\lambda}} = \frac{C_1}{C_0}\sqrt{\left(\frac{\sigma_1}{\sqrt{n_1}\ C_1}\right)^2 + \left(\frac{\sigma_0}{\sqrt{n_0}\ C_0}\right)^2} \qquad (4.1)$$

where C_0 is the number of counts observed at the surface of the filter matrix, C_1 is the number of counts observed at the one inch depth in the filter matrix, σ_0 and σ_1 are the standard deviations in C_0 and C_1; n_0 and n_1 are the number of observations of C_0 and C_1, respectively. The effect on λ of this statistical variability in $e^{-\lambda}$ was evaluated. One standard deviation in $e^{-\lambda}$ was first added to and then subtracted from the $e^{-\lambda}$ value to yield a lower and an upper limit of $e^{-\lambda}$. The difference in λ values corresponding to these limits is, therefore, the range of two standard deviations in λ and for most filter runs fell within the range 0.02-0.05. Many runs were duplicated and the corresponding calculated λ values usually differed by no more than 0.04, which is within the calculated expectation of variability. Uncertainties of this order of magnitude are not significant in determining relationships such as those shown in Figures 28 through 31.

Effects of Dissolved Activity

The effects on filter effluent and the filter medium due to passage of dissolved activity through a filter were investigated. A sudden influx of dissolved radioactivity was caused by pouring a solution of

cesium-137 chloride into the influent of the experimental filter which contained 20-30 mesh sand in order to simulate the sudden arrival of a slug of radioactivity such as might occur following an upstream accident. The resulting activity was monitored by two Geiger counters, one placed inside the filter bed and one immersed in the filter effluent line. Readings were recorded continuously.

Figure 27 shows the results after the background was subtracted and the data were normalized to a constant area under the curves. Curve A illustrates the rise and fall of the activity level within the sand bed, and curve B gives similar information on the filter effluent. From the initial background level denoted by zero on the activity scale, the activity level in the sand (curve A) is seen to rise very abruptly as the radioisotope begins to enter the sand. A sharp maximum is reached in less than 30 minutes, and thereafter the curve decreases rapidly for a time and then at a more gradual rate. The activity becomes almost constant after six hours.

Activity of the effluent (curve B) follows a somewhat similar pattern, but at a time later than that of the sand. The delay in arrival of activity at the filter exit is due to 1) the time required for liquid to pass through the filter and 2) the capacity of the filter to sorb and retain activity. Sorption by the sand is proportional to concentration of dissolved matter in the liquid, so that a portion of any entering activity will be retained so long as the concentration is increasing and the storage capacity of the sand is not exceeded. If the effluent concentration remains constant, an equilibrium is established and no further changes take place.

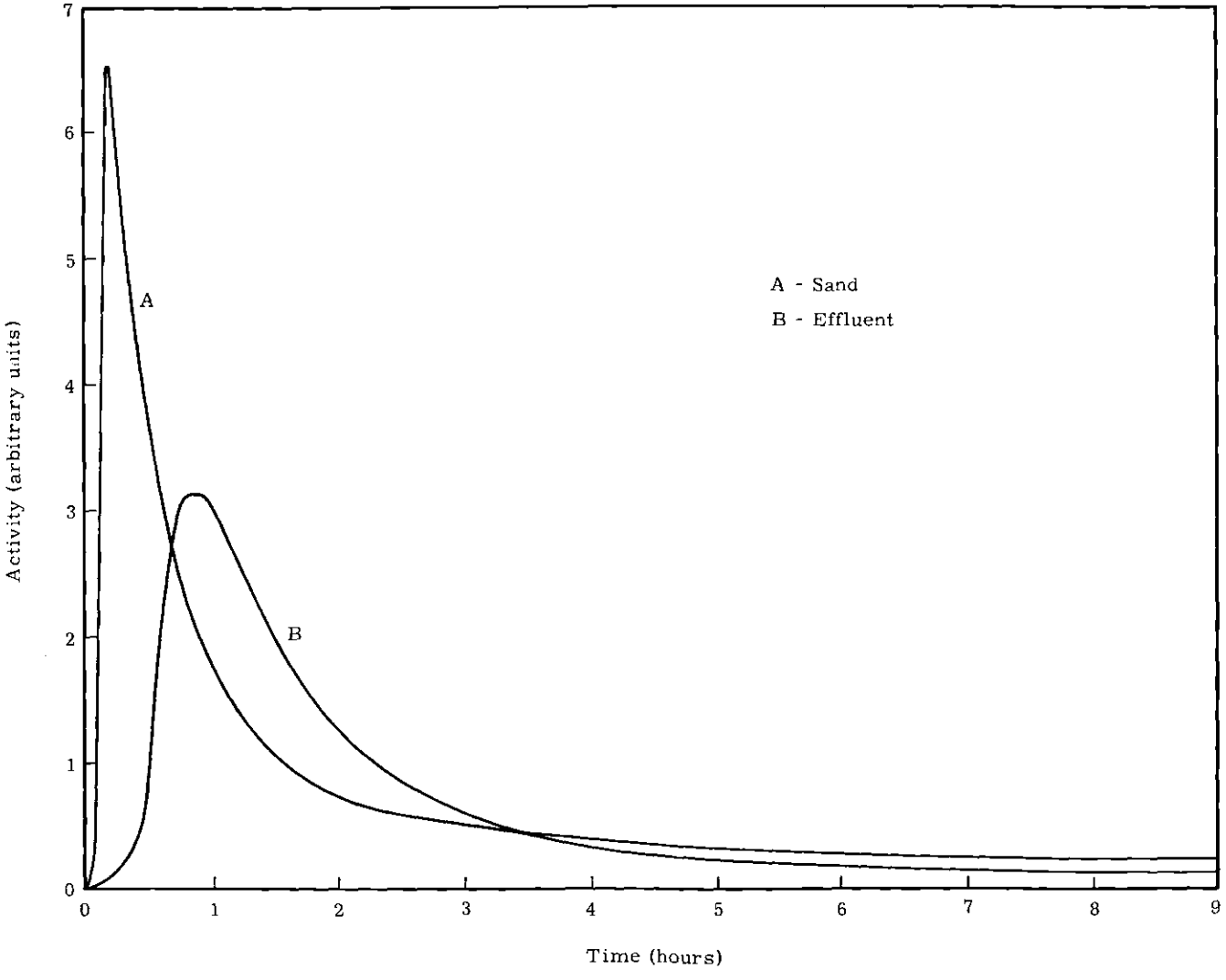

Figure 27. Time-activity Relationships for Effluent and Filter Bed

The effects of dissolved activity on different bed materials was observed in another test where samples of the filter media were exposed to dissolved activity resulting from the presence of uncalcined labeled particles.

A 1.5 gram sample of 88-105 μ vermiculite was stirred for two hours with Cs-137 solution, and another sample was treated in the same manner with I-131 solution. In each case, the particulate matter was then washed repeatedly by stirring with tap water and decanting.

The particles were stirred 30 minutes with 10 grams of washed Anthrafilt No. 1 in about 500 ml of water. The mixture was then allowed to stand approximately 70 hours. A sample of the supernatant was removed, the Anthrafilt washed free of visible vermiculite particles, and five ml portions of both the supernatant and the Anthrafilt were counted in a well-type scintillation crystal.

Under the conditions of this experiment, the activities of the wash waters were very similar, and the final count rates of the supernatants were essentially identical. Under equilibrium conditions, however, the Anthrafilt had sorbed almost 17 times more Cs-137 activity than I-131 activity. Actual values are shown in Table 10.

In another experiment, the affinity of the Anthrafilt for the I-131 was quantitatively determined. A 40 gram sample of washed Anthrafilt No. 1 was put into a beaker containing about 500 ml of water. Iodine-131 was added, the mixture stirred for an hour and allowed to stand overnight. The supernatant was decanted and the activity of a sample of the Anthrafilt was determined with the well crystal counter. The Anthrafilt was then washed by vigorous stirring with tap water,

allowing it to stand several minutes, and decanting the supernatant. The activity of the Anthrafilt was measured several times, and the results are shown in Table 11.

Table 10. Isotope Sorption by Anthrafilt (gross counts per minute)

Sample	Cs-137	I-131
Final wash water	565	157
Supernatant	6,050	5,946
Anthrafilt	267,427	16,412

Table 11. Activity of Anthrafilt Exposed to I-131 (gross counts per minute)

Before Addition of I-131	135
After Addition of I-131	3,508
After 4 Wash Cycles	1,830
After 7 Wash Cycles	1,189
After 10 Wash Cycles	606

Summary of Filtration Runs

The calculated values of the filter coefficient were plotted against the arithmetic mean of the particle diameters, and the results are shown in Figures 28, 29, 30, and 31. The information from these figures is central to the purpose of this study, but it is difficult to visualize its physical significance when presented in this fashion. Consequently, Figures 32, 33, 34, and 35, consisting of linear plots of total counts, C, versus position in the filter bed for representative particle sizes were prepared. These figures represent the actual pattern of deposition of suspended matter within the filter and illustrate the effects of particle size on activity profiles. In each case, the greatest penetration of the bed was by the particles of smallest size, and penetration decreased with progressively larger particles. In Figure 32, the profile produced by all particle sizes larger than 25 μ is approximated by the curve for 53-63 μ particles. Plots of the other profiles were omitted for the sake of clarity. The same situation prevails in Figures 33 and 35 where the 25-37 μ curve represents all larger particle sizes. There were greater differences between the profiles produced by different particle size ranges in the 14-20 mesh sand illustrated in Figure 34. Those omitted from this figure were in the approximate region of the 44-53 μ and 63-74 μ curves.

When comparison between figures is made, it is found that the profile for a given particle size range is not constant but is subject to variation. This difference is due to differences in the filter bed grain size. These findings were expected, as there is no apparent reason that porous media with different characteristics should yield iden-

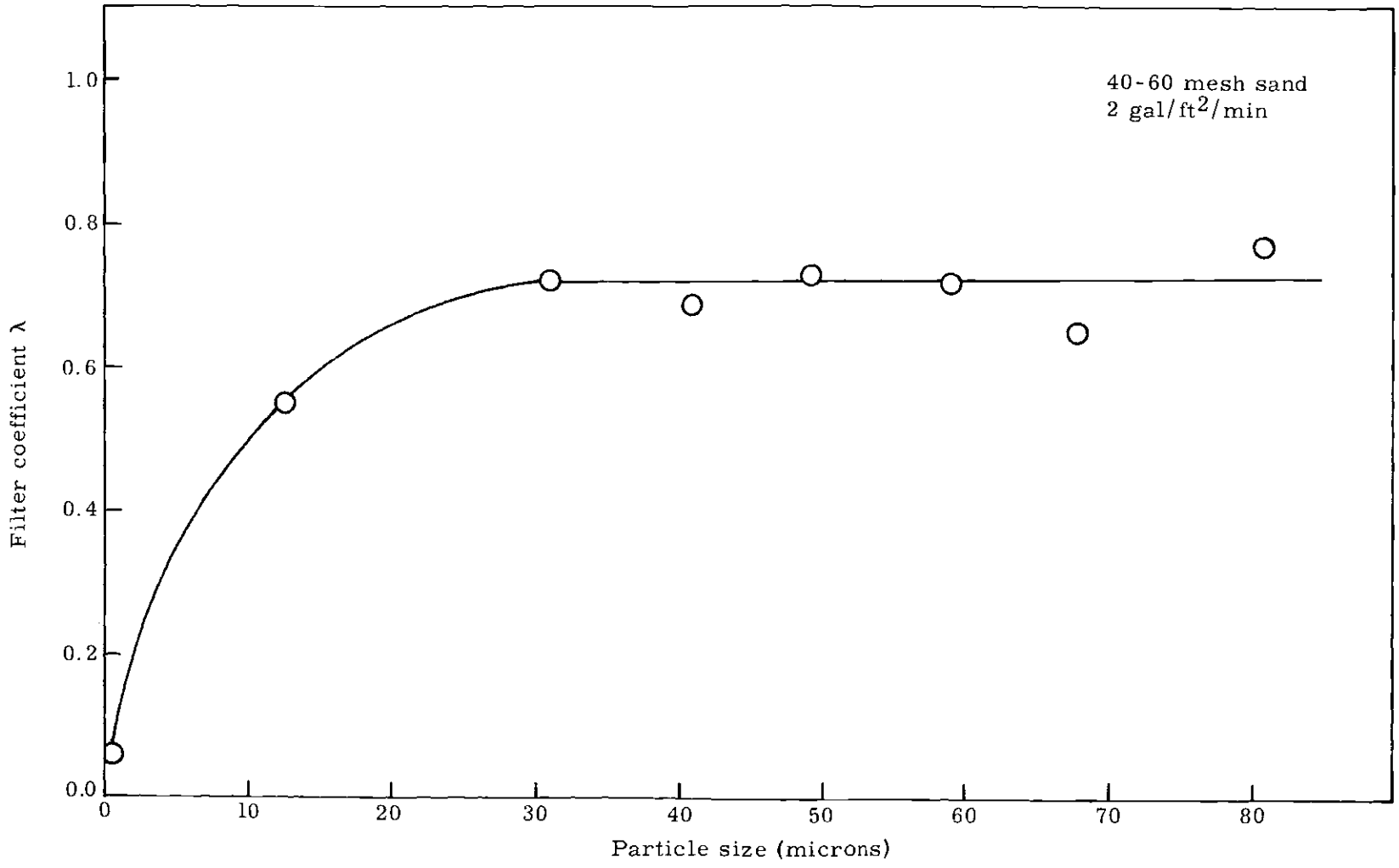

Figure 28. Filter Coefficients for Vermiculite Particles in 40-60 Mesh Sand

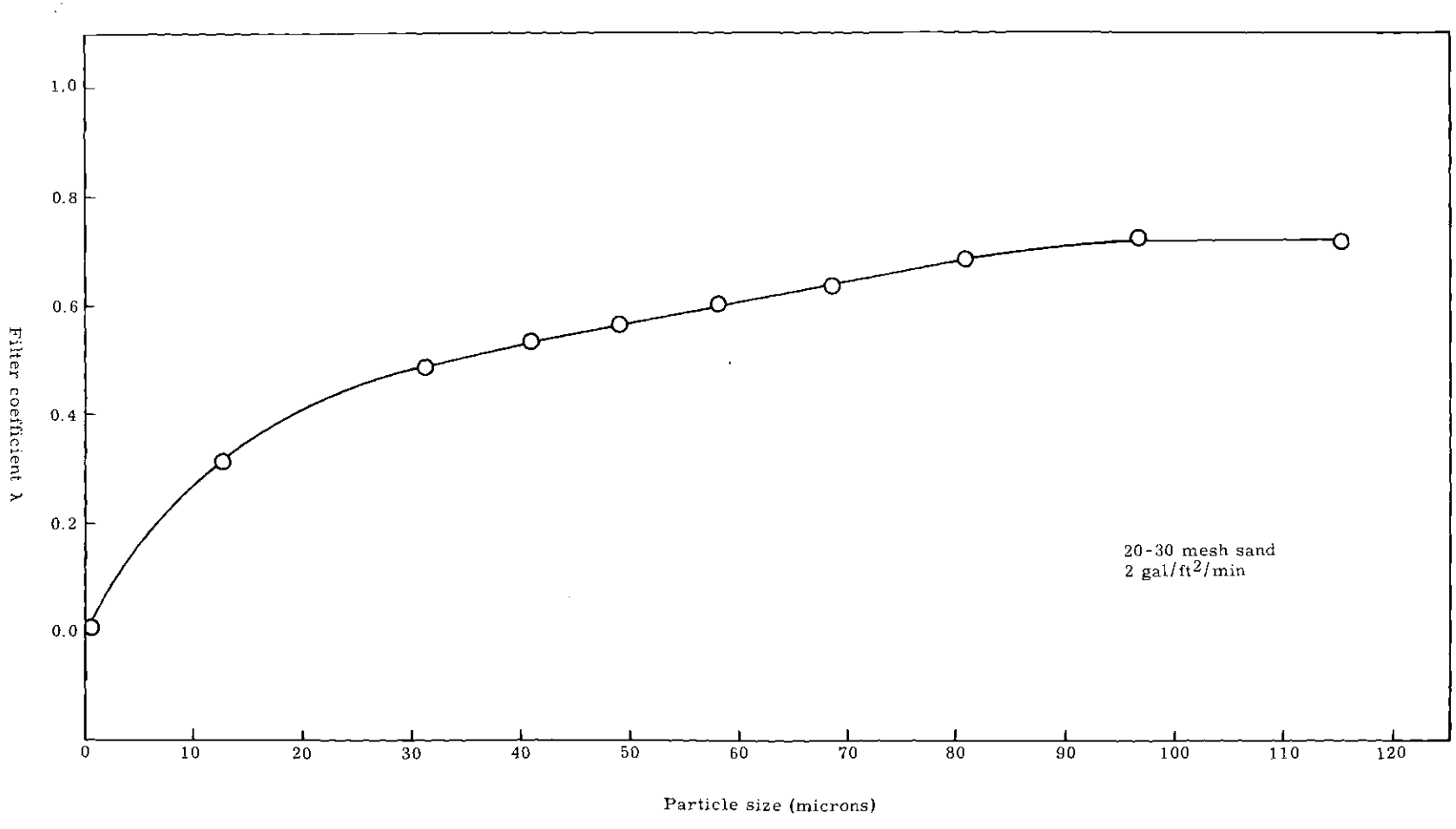

Figure 29. Filter Coefficients for Vermiculite Particles in 20-30 Mesh Sand

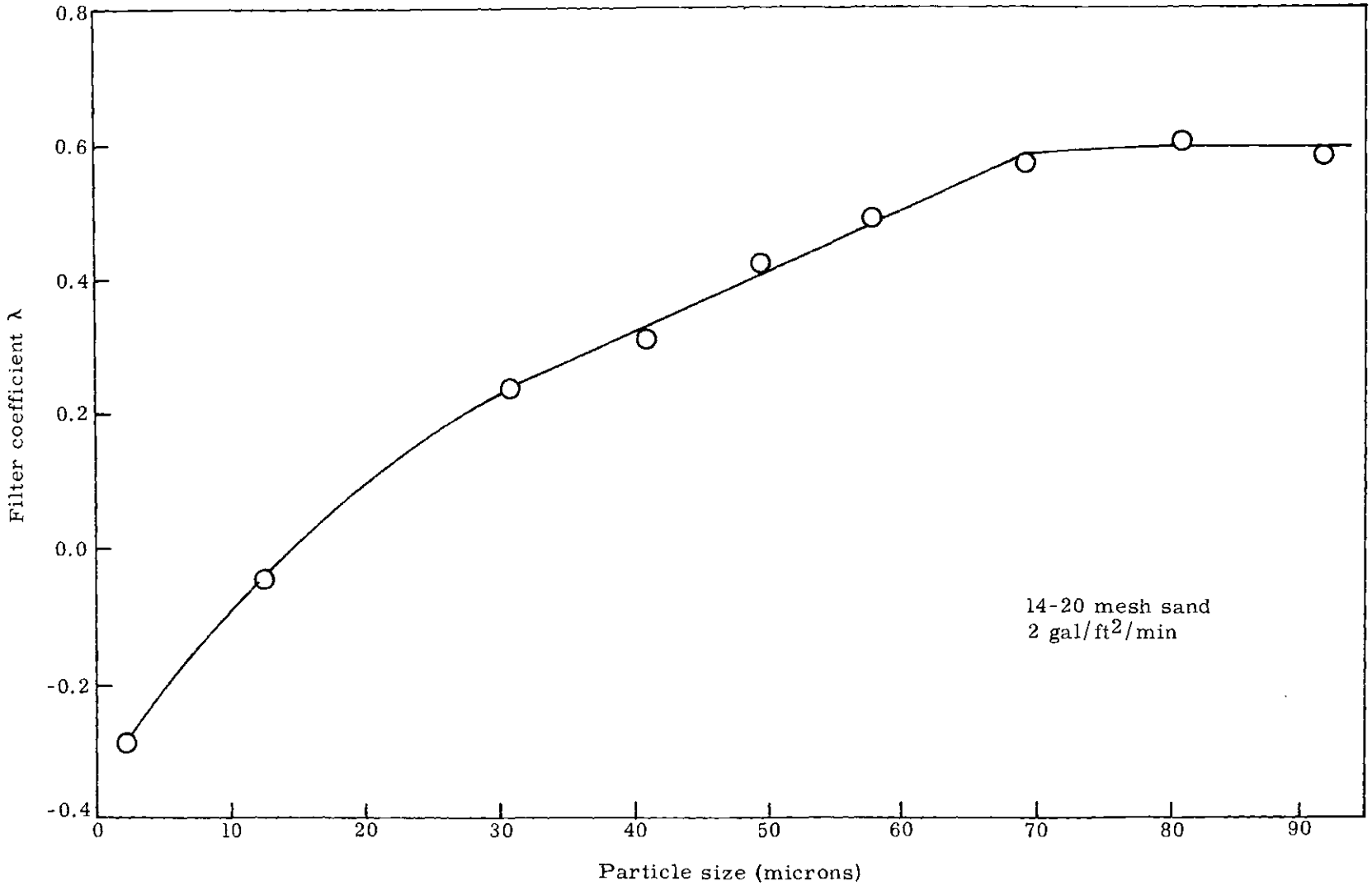

Figure 30. Filter Coefficients for Vermiculite Particles in 14-20 Mesh Sand

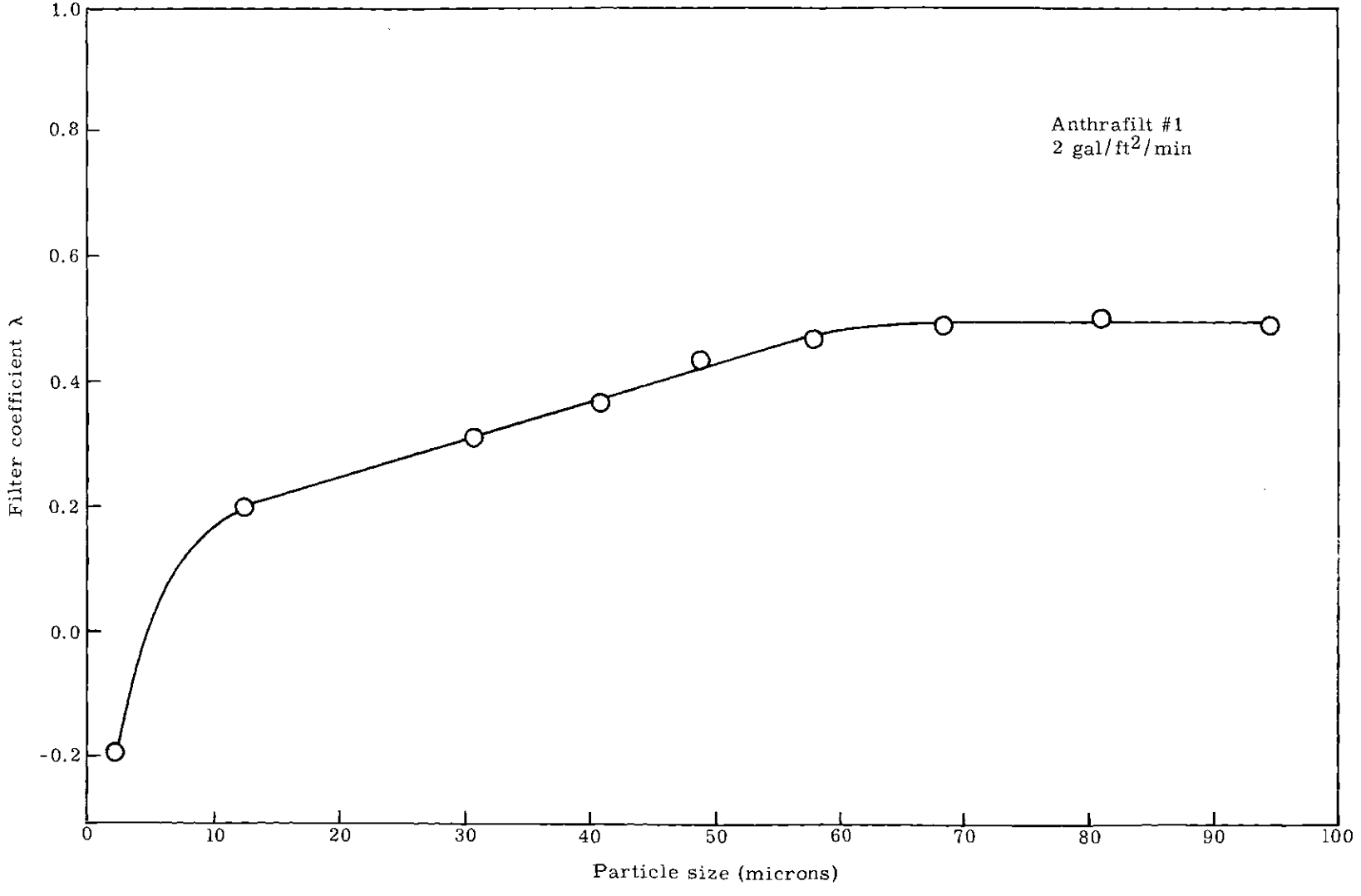

Figure 31. Filter Coefficients for Vermiculite Particles in Anthrafilt No. 1

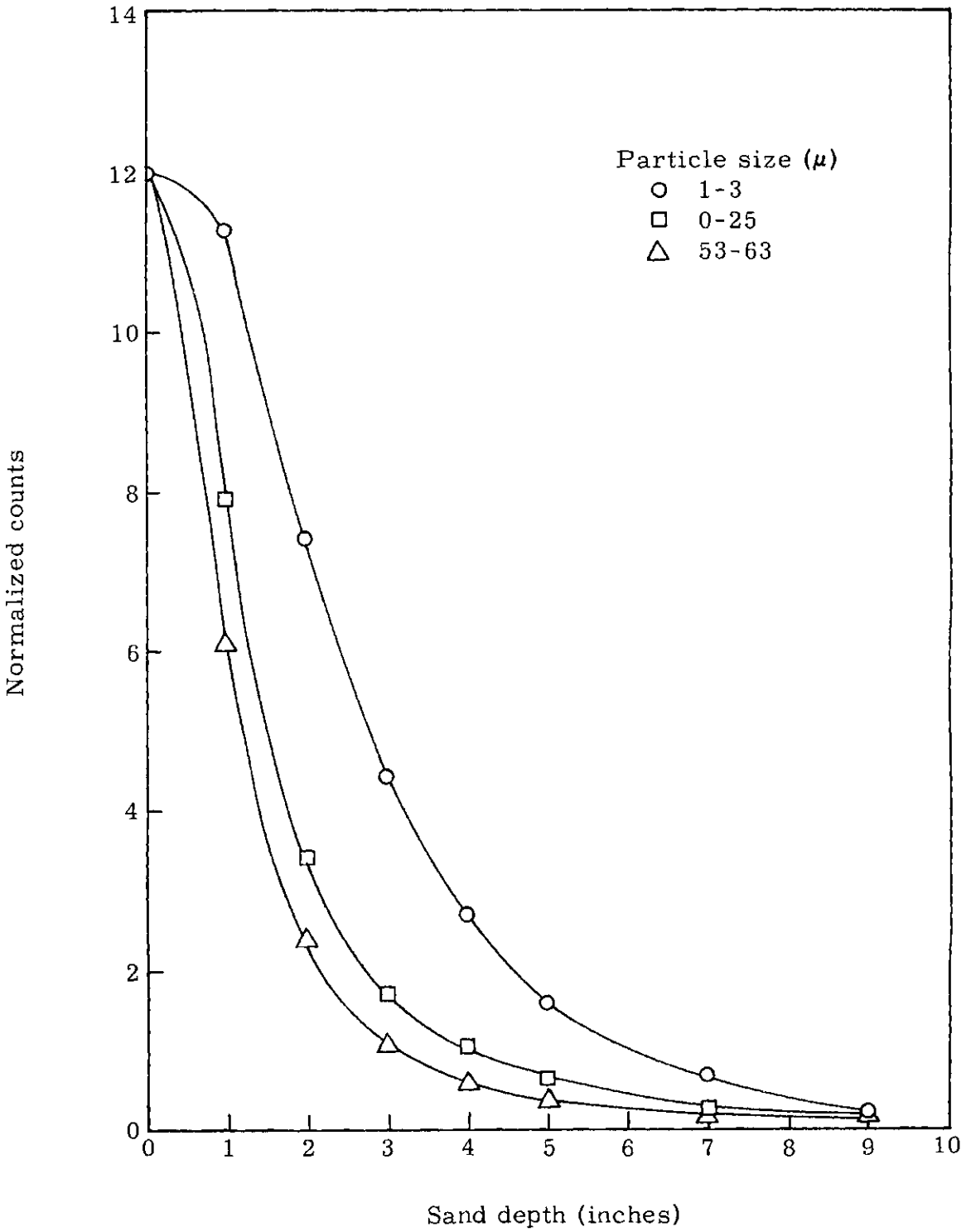

Figure 32. Activity Profiles in 40-60 Mesh Sand at 2 gal/ft^2/min

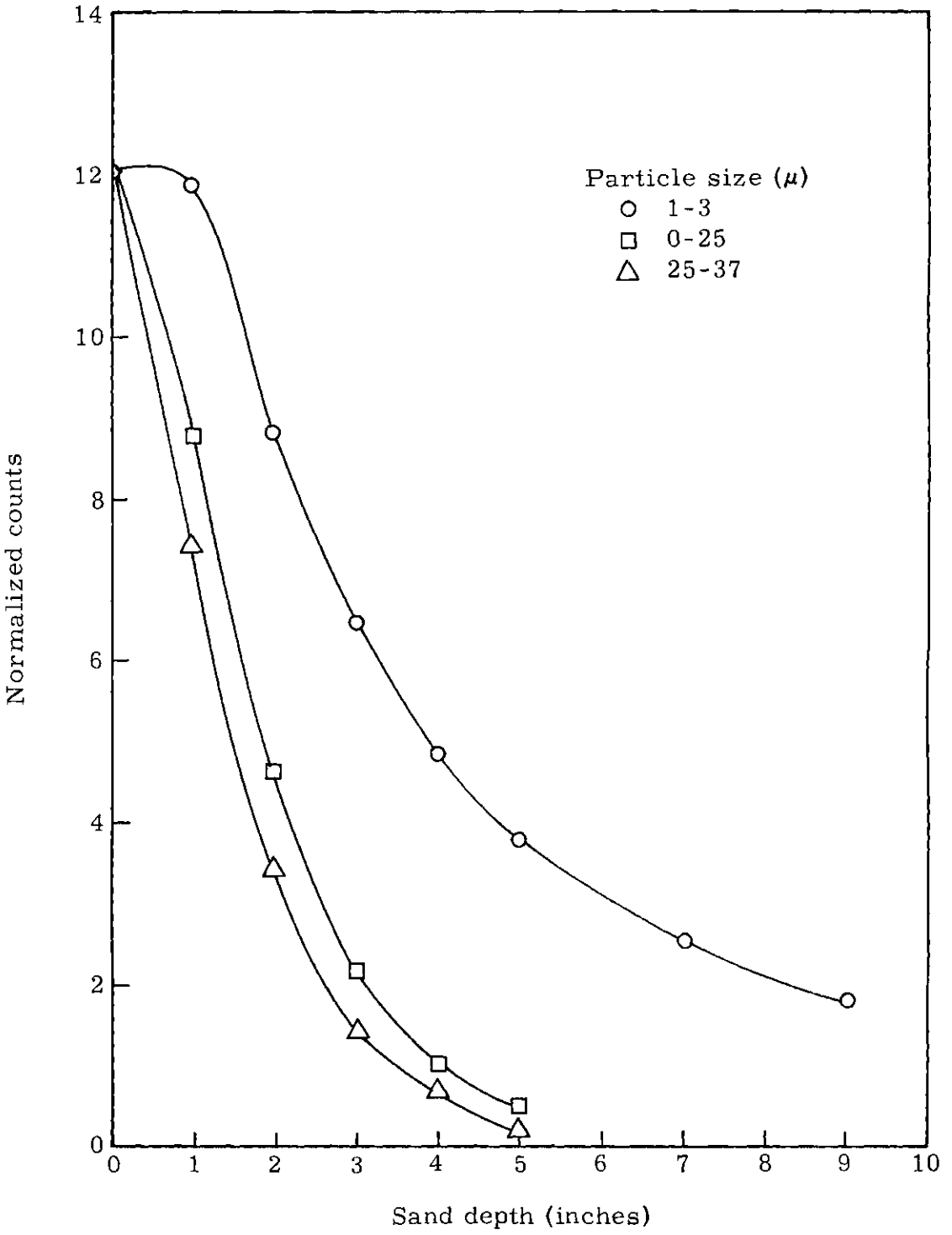

Figure 33. Activity Profiles in 20-30 Mesh Sand at 2 gal/ft^2/min

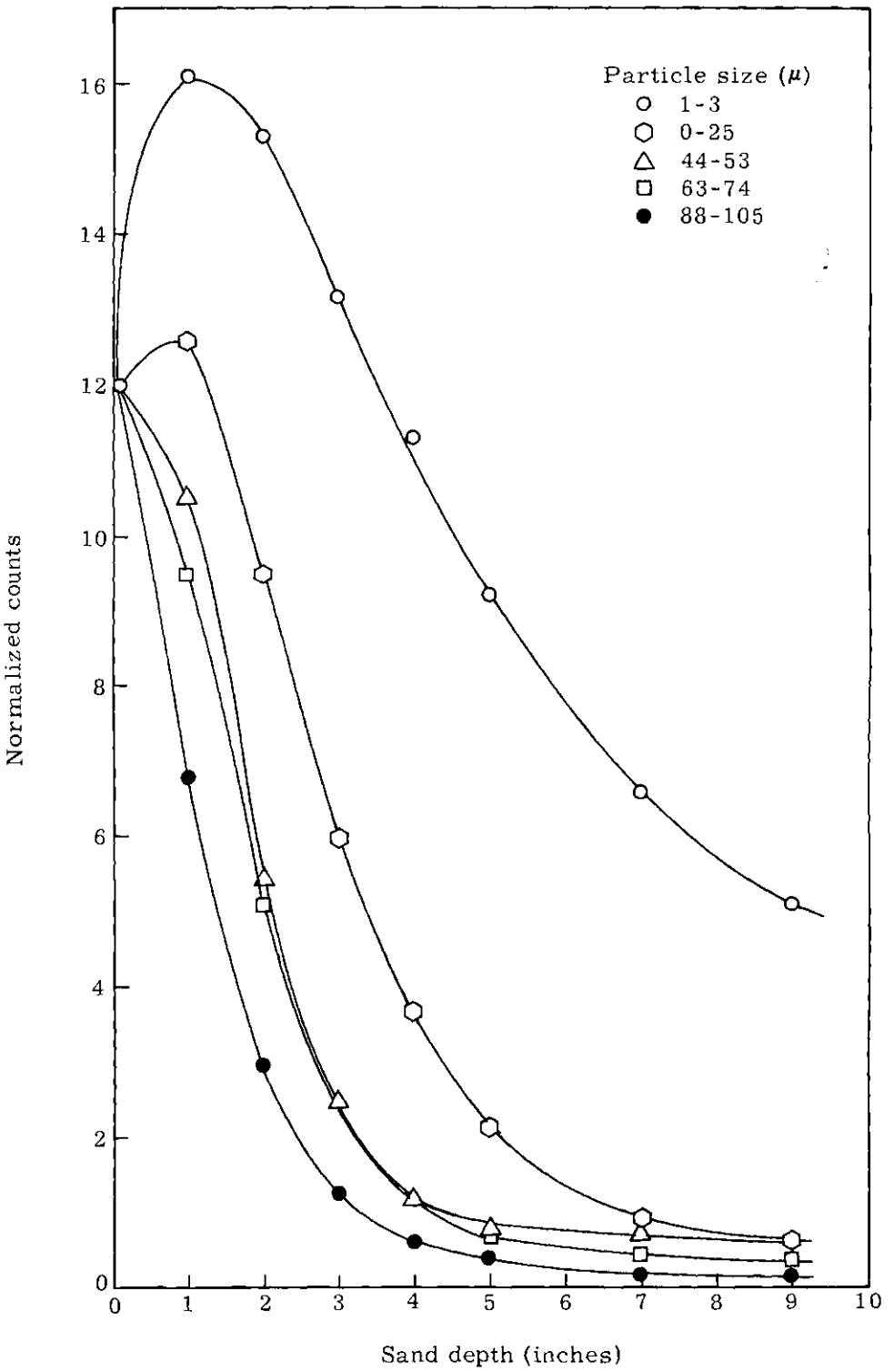

Figure 34. Activity Profiles in 14-20 Mesh Sand at 2 gal/ft^2/min

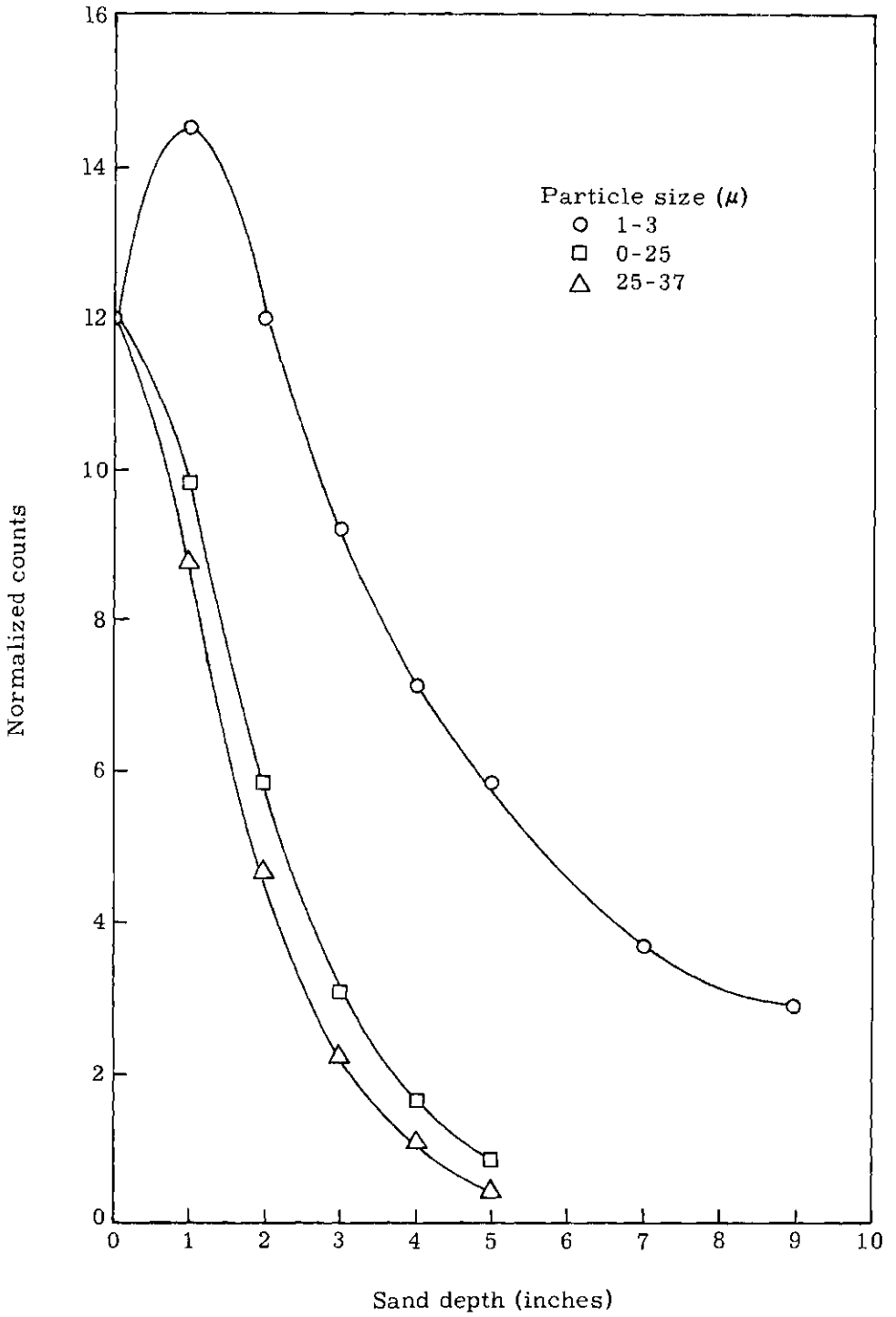

Figure 35. Activity Profiles in Anthrafilt No. 1 at 2 gal/ft^2/min

tical profiles even if the particles involved are the same in each case; however, in view of the similarities of the mechanisms, curves of comparable character would be expected.

If particle deposition is exponential with depth as is generally assumed, the profile curves should plot as straight lines on a semi-log grid. Accordingly, semi-log plots were prepared for a number of different particle size ranges in each medium, and these are presented in Figures 36 through 39. In most cases, these curves are essentially linear for depths of one to five inches. The lack of linearity in the first inch of depth, particularly for the smallest particles, is attributed to geometrical considerations involved where there is much penetration. Below the five inch depth the actual count rate was quite low and was about the same order as the background. Little significance can therefore be attached to the values obtained at the lower levels of the filter. The expectation of an exponential pattern of removal, at least over a specific range, is therefore confirmed.

The time relationship of observed counts was examined in plots such as Figure 40, where the total number of counts at various levels is shown as a function of time. The random fluctuations around the steady level shown by these values are well within the range of normal statistical variation. In a few isolated instances there would occur a set of observations in which the values at all levels would deviate in the same direction from preceding and/or succeeding observations. Such cases were attributed to possible external or environmental influences such as drift of the high voltage or changes in water temperature. As the only significance of these values is in their relative magnitude,

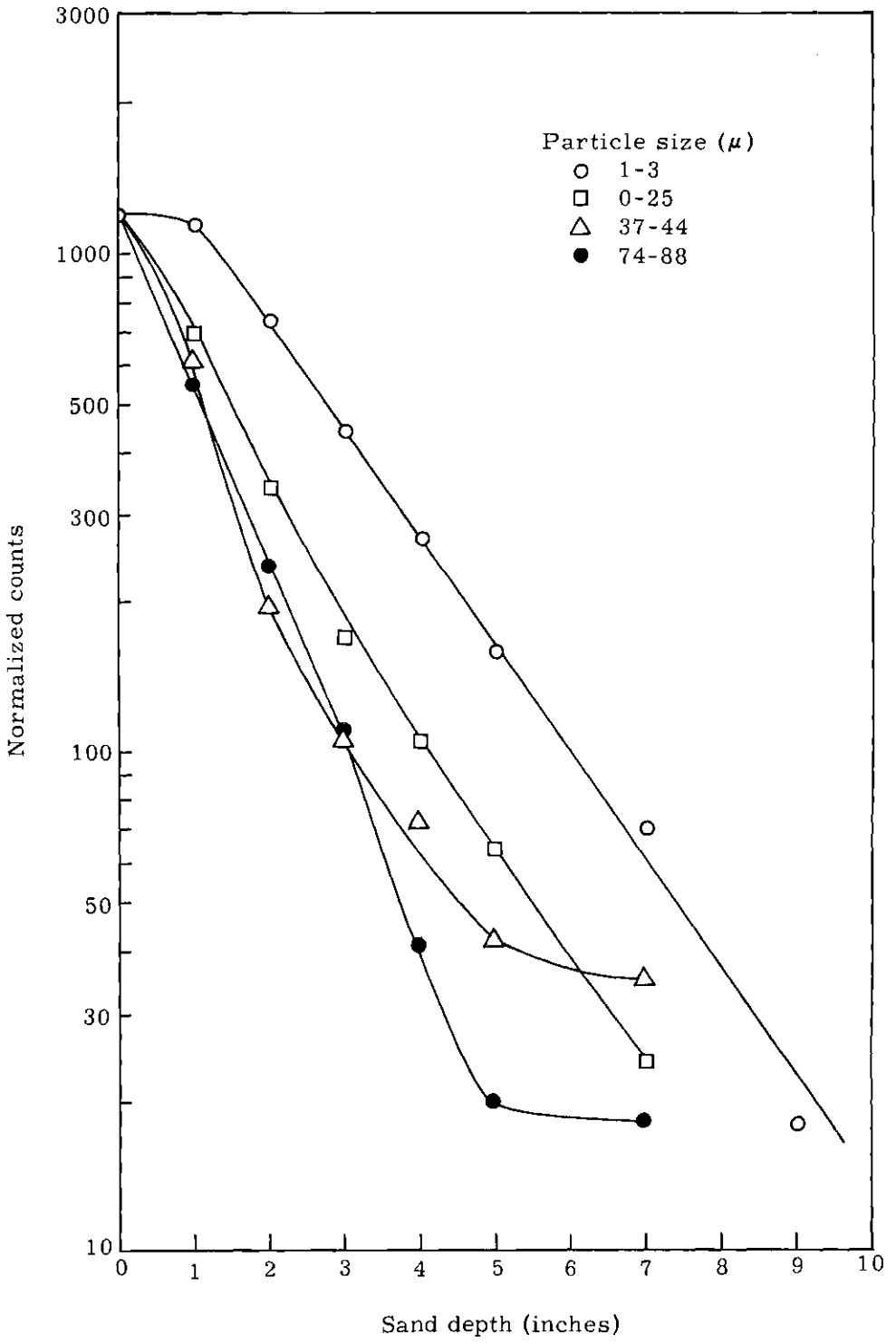

Figure 36. Semi-log Plot of Activity Profiles in 40-60 Mesh Sand at 2 gal/ft^2/min

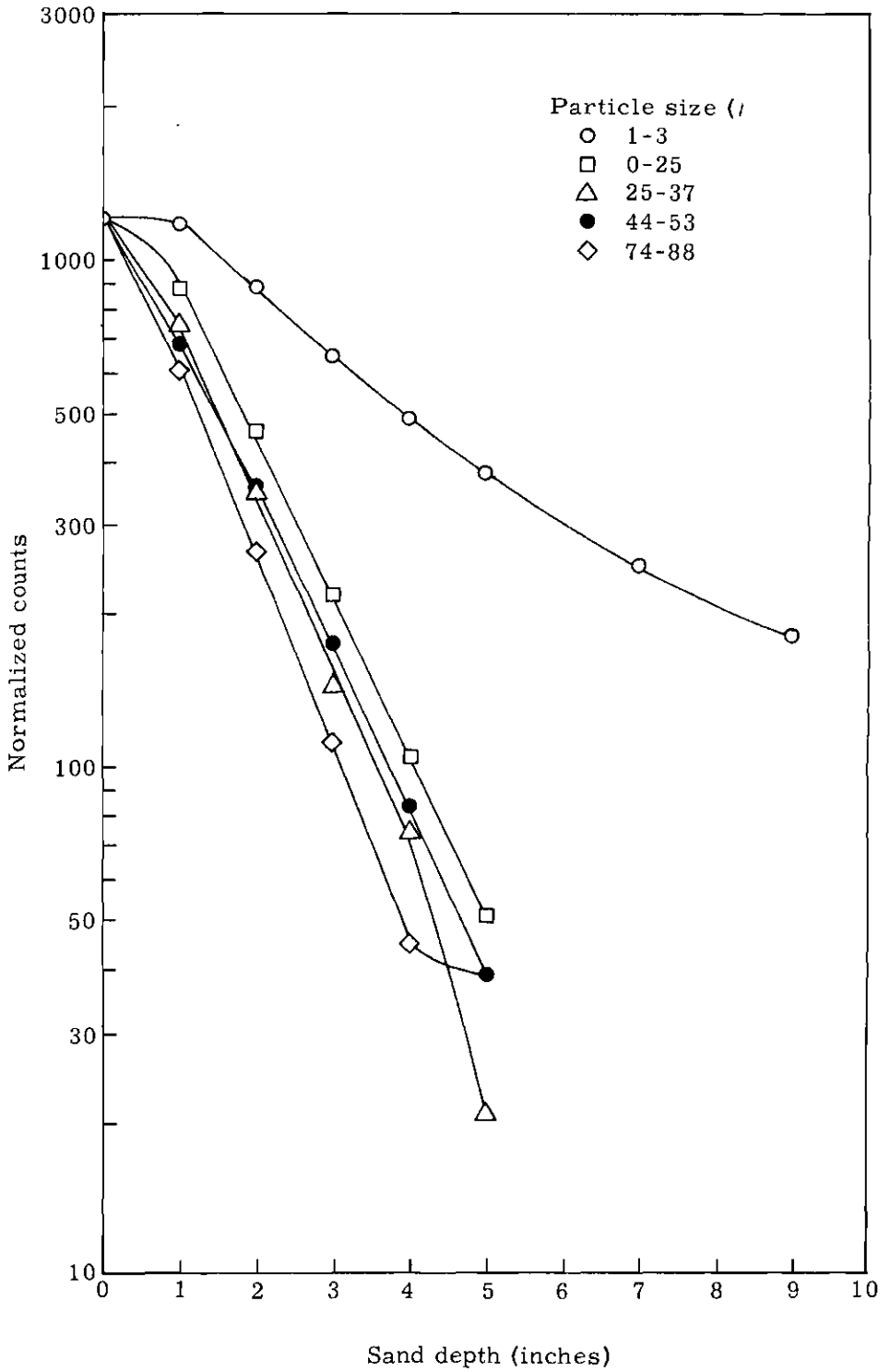

Figure 37. Semi-log Plot of Activity Profiles in 20-30 Mesh Sand at 2 gal/ft^2/min

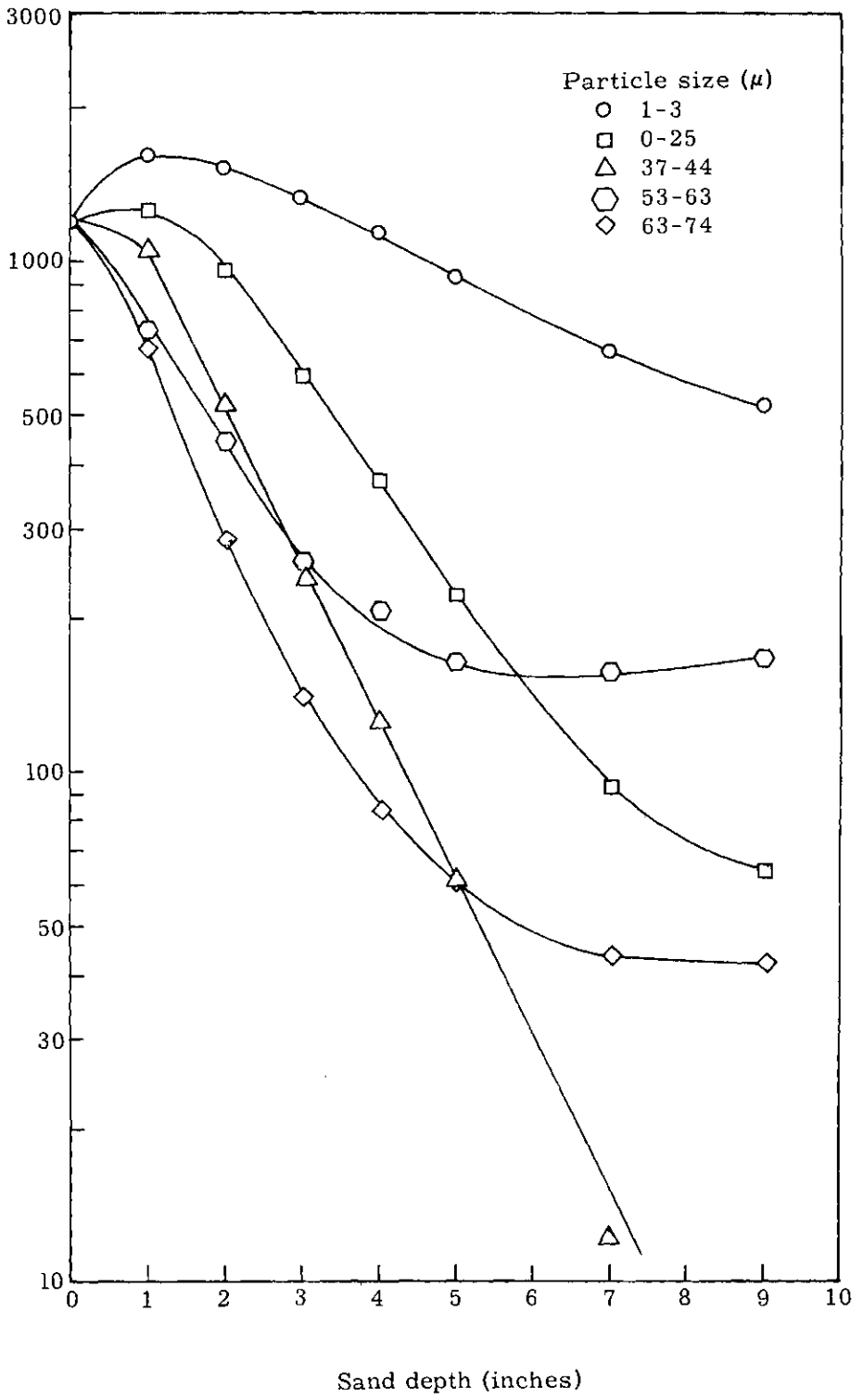

Figure 38. Semi-log Plot of Activity Profiles in 14-20 Mesh Sand at 2 gal/ft^2/min

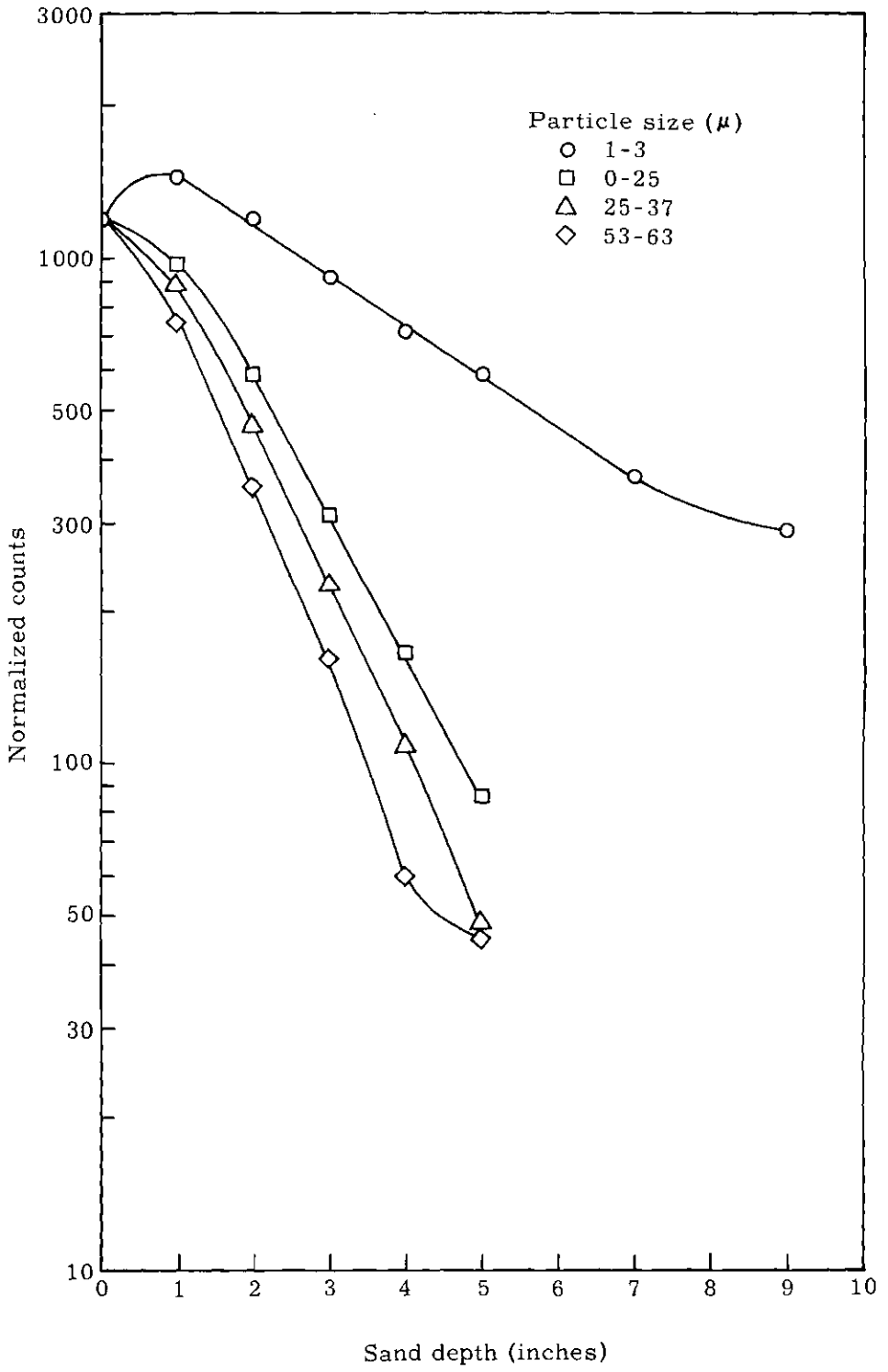

Figure 39. Semi-log Plot of Activity Profiles in Anthrafilt No. 1 at 2 gal/ft^2/min

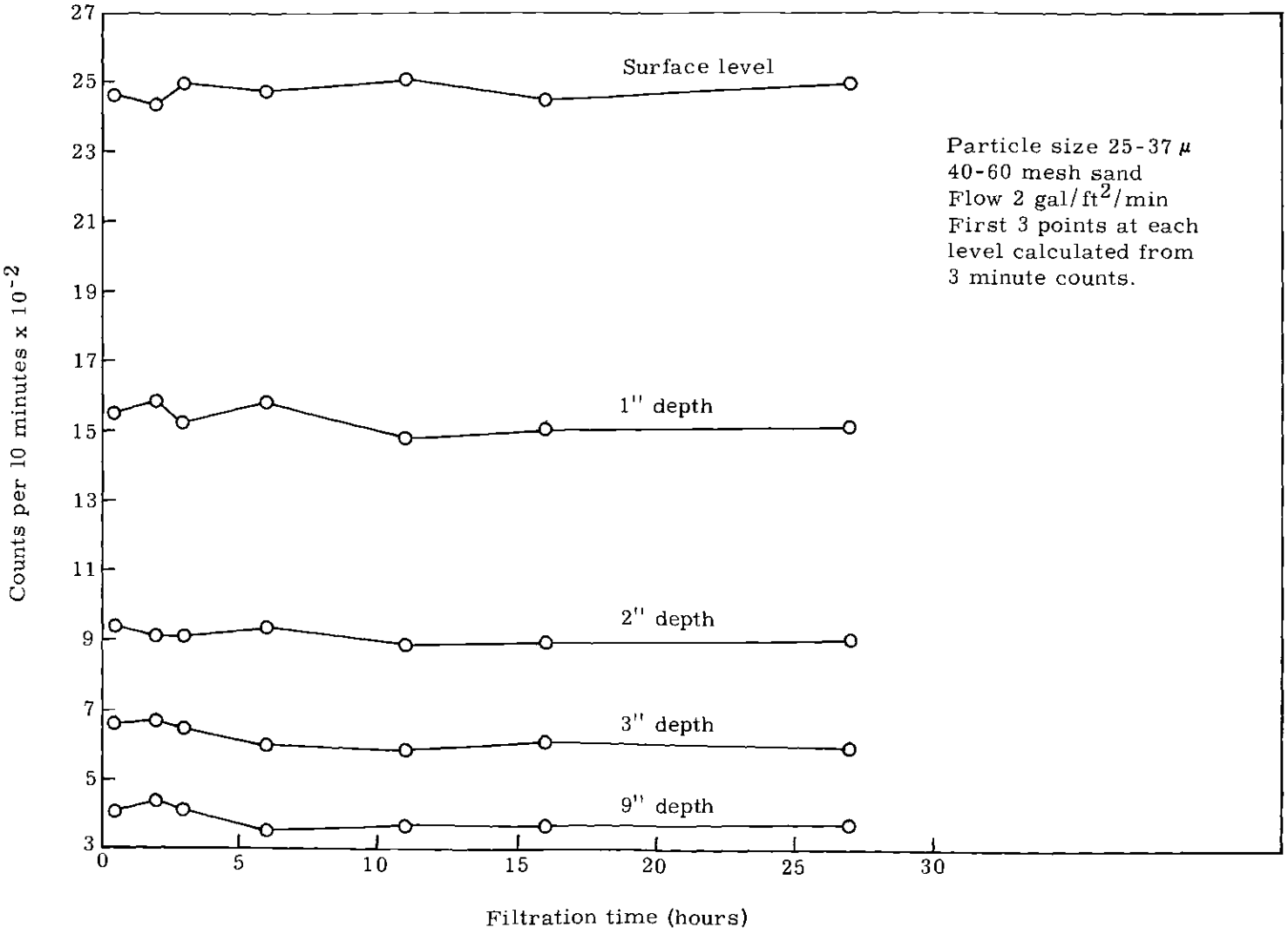

Figure 40. Typical Count Variations at Different Sand Levels

variation of a whole set of readings was of little significance where the ratio of values remained essentially constant.

The exact relationship of bed grain size and filter coefficient has been the subject of discussion in the literature. The data obtained in this study were consequently plotted in a manner so that this relationship could be examined. In Figure 41, it may be seen that this relationship depends to a considerable degree on the particle sizes involved. The only straight lines observed are for the 63-74 μ and 74-88 μ particles, and the latter is questionable due to an exceptionally high value (0.78) for λ in the 250 μ (40-60 mesh) sand. Referring to Figure 28, it seems more likely that this value should be about 0.72, which would produce the dotted line included in Figure 41. The filter coefficient as measured in this study therefore appears as a linear function of sand grain diameter only for 63-74 μ particles. A severe limitation is imposed on the interpretation of these plots due to the availability of only three points for each curve, but it is clear that the size of the suspended particles is involved.

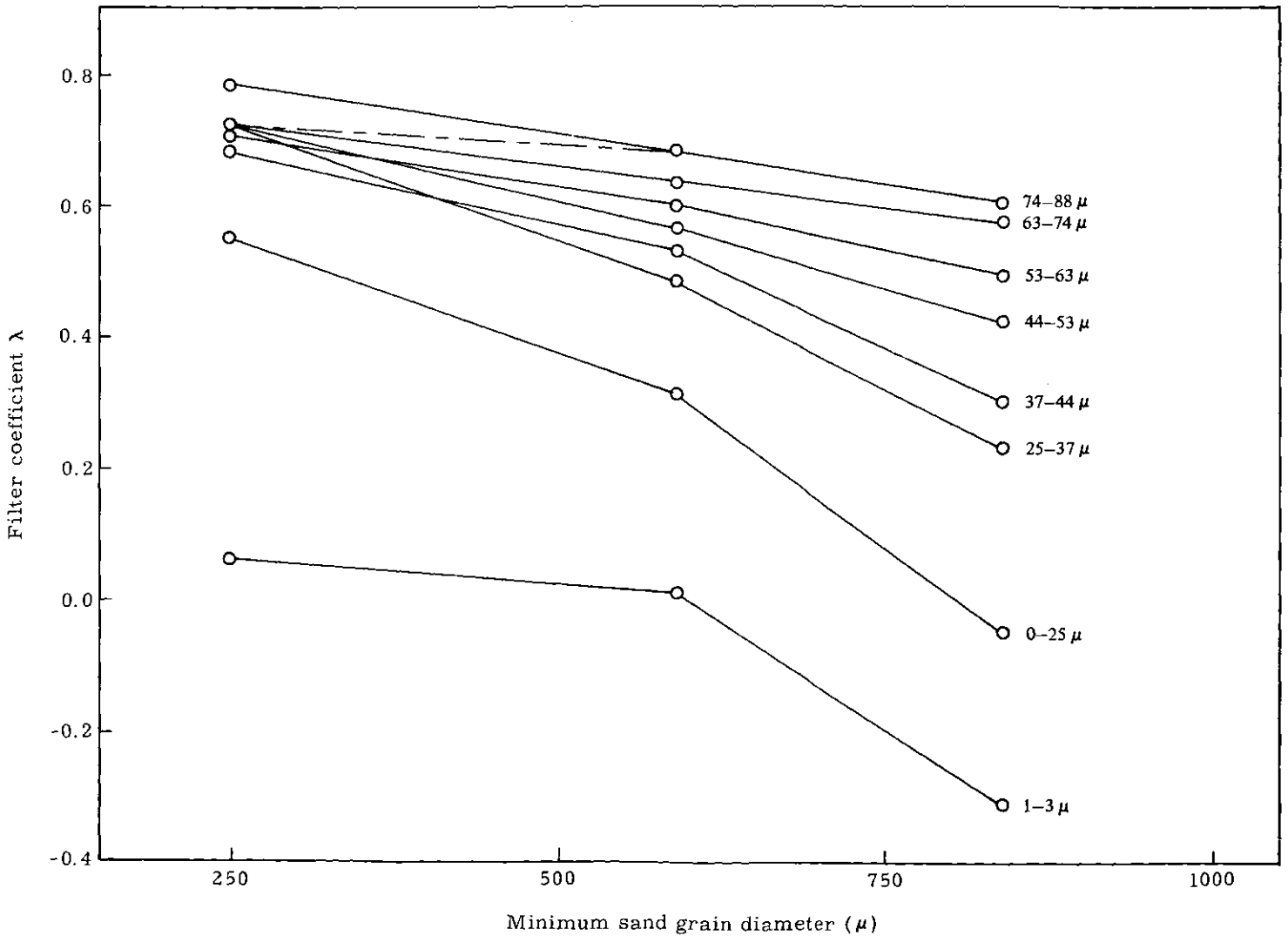

Figure 41. Filter Coefficients for Minimum Sand Grain Diameters

CHAPTER V

DISCUSSION OF RESULTS

In the following discussion, both the suspended particles and bed grains are treated as though they are uniform spheres, but it should be clearly understood that this is not the actual case. The solids are somewhat angular and vary slightly in size, and the interstitial void spaces are not the geometrically uniform shape that would be produced by regular packing of spheres. Nevertheless, there seems to be no practical alternative to consideration of things as though they are ideal and then relating the results back to physical reality.

The filter coefficient has a definite physical significance as a measure of filter operation; it is directly proportional to the percentage of suspended matter removed by each layer of the filter. A high value indicates that suspended matter will be deposited in such a fashion that the top layer receives much more material than lower layers which will trap successively smaller amounts. A value of zero for the filter coefficient would indicate that the concentration of suspended matter in the effluent is equal to the concentration in the influent; that is, no filtration is occurring. A situation encountered in some instances in this study involved deposition such that the concentration of deposited matter was greater at the one inch depth than at the surface. The resulting filter coefficient calculated from such occurrences has a negative value. Physically this would mean that the filter is

adding rather than removing suspended matter, and this is obviously not the case either here or elsewhere. This inversion of deposit concentration only occurs in a fraction of the filter depth, as the coefficient over the entire depth must be equal to or greater than zero. It is therefore to be carefully understood that the negative coefficients encountered in this investigation refer only to the relative concentrations at the points employed to compute the coefficient.

In full-scale rapid sand filtration practice, a coagulant, most commonly alum, is mixed with the incoming raw water which then passes into a basin for a period of sedimentation before reaching the filters. Because of this pre-filter treatment, the suspended matter entering the filter is almost entirely floc, which is a hydrated aluminum oxide-hydroxide complex of variable composition formed by hydrolysis of the alum. The gross physical form of this complex is highly dependent on the quality of the raw water, alum concentration, manner of addition, stirring rate, and other factors, but in general it is a low density, feathery material. This floc traps suspended particles both mechanically and through chemical interaction. The net result is an agglomeration of suspended matter into larger, more rapidly settling agglomerates. The most notable physical characteristic of floc is the ease with which it can be torn apart. The individual flocs are quite fragile and must be handled cautiously to avoid regions where high shear forces will tear them apart.

There is much difference between such soft, low-density floc and the hard rigid vermiculite particles employed in this study. Experimentally it is quite difficult to obtain a reproducible floc of uniform

particle size because of the many variables that determine this property at initial formation and the likelihood of subsequent change due either to forces of attrition or coalescence. With rigid particles, it is unlikely that any change in size will occur during the course of the experiment. As this study has focused primarily on particle size, the differences between floc and vermiculite were accepted for the sake of assurance of a constant particle size.

This investigation has been concerned with events that occur at the beginning of a filtration run when the bed is free of trapped matter. As previously discussed, certain characteristics of the bed as evidenced by the filter coefficient may change as matter is deposited, and this may possibly alter the relative importance of the various mechanisms. Although appreciable change seems unlikely, it is necessary to consider the results obtained here as strictly applicable only to clean bed conditions.

Considering the sand grains as regularly packed spheres of uniform size, it can be shown that the maximum diameter sphere that could pass through the interstices of the pack is 0.155 times the diameter of spheres comprising the pack. The calculation is as follows. As shown in Figure 42, the centers of the large spheres form an equilateral triangle; angle CAB is therefore 30°. Then $\overline{AB}$ = R sec 30° = 1.1547 R. r = $\overline{AB}$ - R = 1.1547 R - R = 0.155 R and the diameter of the small sphere is 0.155 times the diameter of the large. As the sand employed actually varies over a range of sizes, for calculation purposes the diameter of the smallest of the sizes present was used for calculation of the theoretical minimum size of the openings between the grains. On this basis,

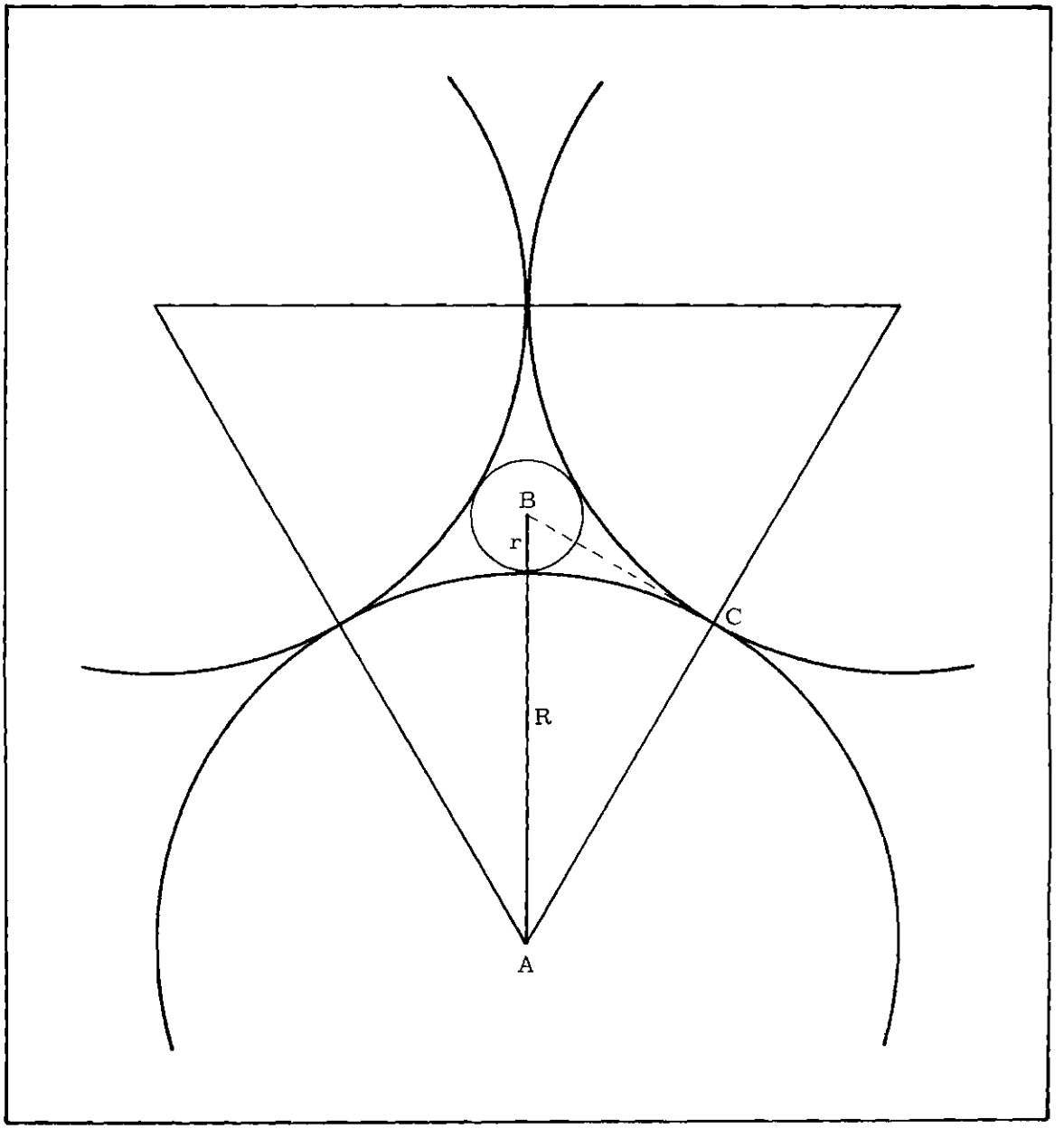

Figure 42. Idealized Geometry of a Pore

the 40-60 mesh sand should pass spheres of diameter less than 38 μ; 20-30 mesh, 91 μ; and 14-20 mesh, 130 μ.

In any filtration process it is obvious that rigid objects larger than the openings in the filter will not pass through due to physical hindrance. Sand filtration is no exception as may be seen by reference to the plots of filter coefficient versus particle diameter, Figures 28 through 31. Considering the data of Figure 28, it is seen that λ is essentially constant at a value of about 0.7 for particle diameters of about 30 μ or greater for void diameters of 38 μ. As λ represents the ratio of radioactivity detected at the surface to that detected at the one inch depth, it should remain constant, as long as all the activity is at the surface level. This is confirmed by the curves of Figures 29, 30, and 31, where the curves are also seen to attain a constant value for λ for large particles. It is interesting to note that, although there is appreciable difference in the sand sizes represented, the curves of Figures 29 and 30 show that λ becomes constant at about 95 μ and 80 μ, respectively.

This can be explained by considering that there is a wider range of sand grain sizes in the 14-20 mesh sand, and the closer packing possible with grains of differing sizes results in a maximum opening somewhat lower than would be expected with unisize grains. However, this must apply only to the maximum opening, as the average pore of the 14-20 mesh sand is undoubtedly larger than the average in the more nearly uniform 20-30 mesh sand. This hypothesis is supported by the magnitude of the maximum λ values in the two different sands. The coarser sand has a maximum λ of only 0.6 while the finer sand reaches a value of almost

0.7. The higher λ value indicates that the finer sand is a more effective filtration medium, a finding which is in keeping with general experience.

With Anthrafilt No. 1, λ becomes constant at about 70 μ, suggesting that the maximum constrictions are smaller than those of either 14-20 mesh or 20-30 mesh sand for comparable particle sizes. This is not unreasonable due to the large variation in size of the Anthrafilt granules. These granules are also less spherical than sand grains and could be described more accurately, perhaps, as somewhat flat platelets. They pack with less void space than spheres, producing the observed result. The Anthrafilt revealed the lowest maximum λ value, 0.5, of any of the filter media studied. For suspended particles less than about 50 μ in diameter, the Anthrafilt exhibited λ values higher than those of 14-20 mesh sand and lower than those of 20-30 mesh sand.

It should be noted in this connection that a high value of λ is not necessarily the most desirable situation in the design of an actual filter. A lower λ value would tend to spread the deposited load farther downward in the filter which should cause loss of head at a somewhat slower rate. On the other hand, the lower λ value would require a greater depth of filter medium in order to attain the same result. As the relationship of the filter coefficient, λ, to size of suspended particle is not the same for sand and Anthrafilt, this emphasizes the importance of evaluation of size, shape, and character of suspended particles as well as cost of operation in the design of a filter.

In Figures 29, 30, and 31, it is possible to identify three different slopes in each curve. Beginning at the smallest particle size,

each curve rises rapidly at first through a region of decreasing slope and then enters a zone of essentially constant slope, which is followed by a plateau of zero slope. Figure 28 is similar, but there are insufficient points to define clearly the shape of the curve for particles smaller than 30 μ in diameter. One may reason by analogy with the other curves that a region of constant slope could be present. Unfortunately, it was not possible to obtain particle fractions of intermediate size so that this region could be explored more fully. This curve is not contradictory to the others, but conclusions as to its exact shape must remain speculative pending availability of suspension particles of suitable size.

In Figures 29 and 30, the curve segments between 30 μ and 80 μ are linear. In Figure 31, a linear region exists between 12 μ and 70 μ. The linearity of these curve segments is significant in the evaluation of one possible removal mechanism, interstitial sieving. Equation 2.4 shows that the filter coefficient should be proportional to the three-halves power of the ratio of diameters of the particles and the grains of the filter medium

$$\lambda \propto (D/d)^{\frac{3}{2}} \tag{2.4}$$

Plots of λ versus $(D/d)^{\frac{3}{2}}$ for 20-30 and 14-20 mesh sand are given in Figure 43, and a plot of the same ratio for Anthrafilt No. 1 is given in Figure 44. Considering only physical hindrance, if the experimental findings are in agreement with the theoretical relationship, these plots should be straight lines. The curve for the 20-30 mesh sand is seen to be linear or very nearly so over a considerable portion of its length,

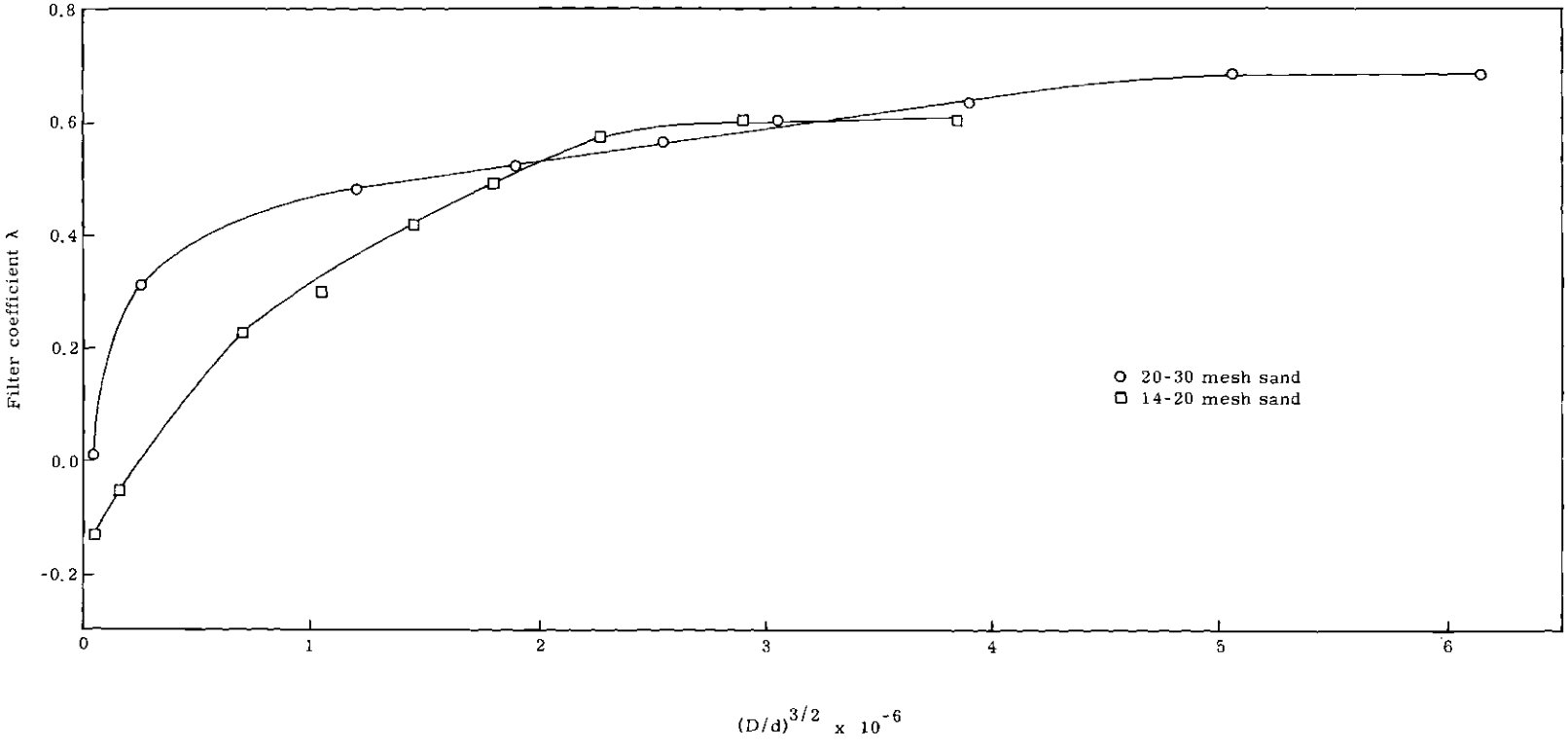

Figure 43. Correlation of Idealized Pore Area Ratio and Filter Coefficient for 20-30 and 14-20 Mesh Sand

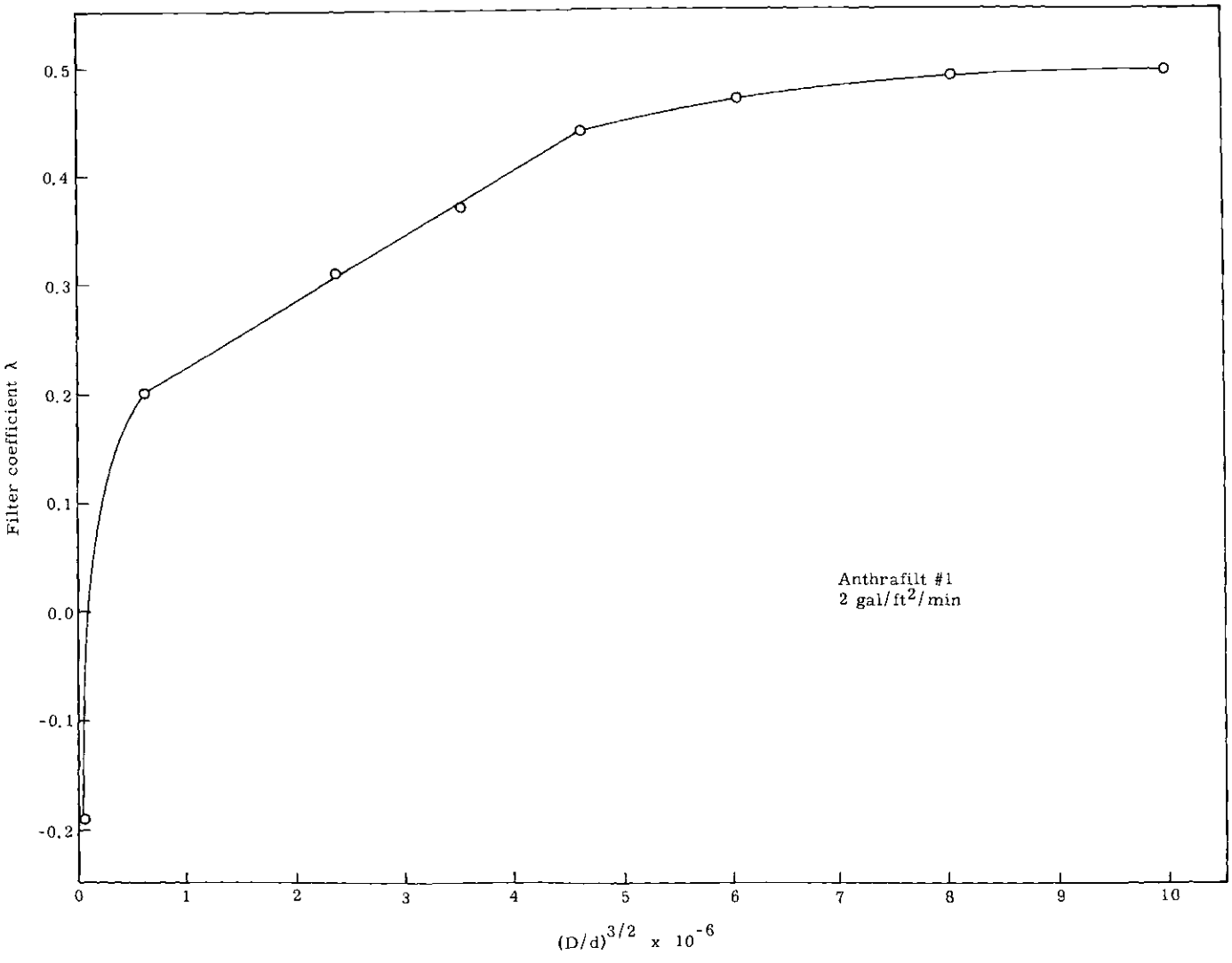

Figure 44. Correlation of Idealized Pore Ratio Area and Filter Coefficient for Anthrafilt No. 1

but it departs rapidly from linearity as D/d becomes very small. The linear portions of the curves for the other two filter media are somewhat shorter and less distinct, but all three curves exhibit the same general characteristics. The transition away from linearity begins in the sands as particle size becomes less than about 30 μ. With Anthrafilt No. 1, the linear region extends down to near 10 μ.

It is, therefore, concluded that interstitial sieving is an operative mechanism, at least over a certain range of particle sizes. It is apparent, however, that other forces are involved and no doubt predominate in the regions where the curve is non-linear. If the sieving mechanism is assigned the notation f_1 in equation 2.1, this implies a high value for "A" for particles with sizes in the linear region.

It has been pointed out above that the flow was laminar, and the velocity profile was therefore parabolic. The situation can be more clearly illustrated if a short section of an individual flow channel through the bed is considered circular. Figure 45 depicts a roughly spherical suspended particle, and the arrows denote velocity streamlines.

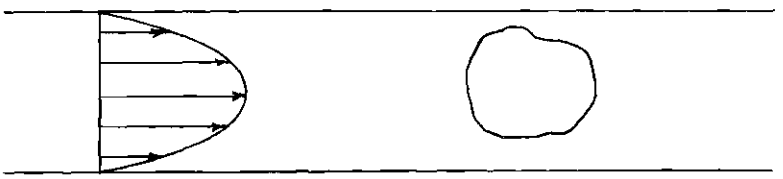

Figure 45. Theoretical Velocity Profile in an Idealized Situation

It can be seen that a particle of the size shown or larger will be subjected for the most part to streamlines of maximum or high velocity. Even if the particle were touching the wall, only a small fraction of its surface would be immersed in the region where flow velocity is approaching zero, and the particle would not be slowed greatly even in this limiting position. The net result is that for particles that are large with respect to bed constrictions the velocity appears to be essentially uniform across the opening. The situation is different for a particle of small diameter which has a possibility of moving along close to the wall in a region of low velocity. For such small particles, flow no longer appears uniform but diminishes from a maximum at the center and approaches zero at the wall.

This being the case, the ratio of flow volume through areas A' and A (equations 2.2 and 2.3, see Figure 4) is not the same as the ratio of the areas but must be modified by consideration of the velocity profile in each area. Where A' is large, the situation where large particles may be trapped, the ratio of areas is a good approximation of the ratio of flows, but this approximation progressively worsens as A' decreases. Flow volume through very small triangles is exceedingly small, little suspended matter can be trapped, and the filter coefficient is low. This is one factor that contributes to the rapid decrease in λ for particles smaller than those appearing in the linear zones of the graphs of Figures 28 through 31.

While size relative to bed constrictions is important, the absolute size of suspended particles is also a consideration, as small particles must be removed from a region of the streamline where the

velocity is very low. This implies that some force other than physical hindrance operates in the entrapment of very small particles, and this is undoubtedly the case. If sieving were the only mechanism, the particle would have to move back into the crevices where the velocity approaches zero, a condition that produces very poor filtration efficiency. Further, one might assume that as filtration proceeds, the smaller crevices would become filled, and only progressively larger particles could be removed. It is well known, however, that small particles continue to be removed even after most of a filter is clogged, and the probability of crevices with small widths remaining unclogged is very low. It is therefore reasonable to assume that it is not necessary to have a particle lodge between two surfaces; a single point of contact is sufficient for particle deposition and adherence.

Figure 46 is a plot of filter coefficient against the ratio of suspended particle diameter to maximum diameter of the sand constrictions. If this ratio were the determining factor, this plot should produce a straight line. Using experimental points for the three different sizes of sand, a line can be drawn by eye through the greatest concentration of points, leaving only a few that appear at considerable distance from the line. In each case, the points farthest away are those of the smallest particle sizes. As previously indicated, the λ values for the smallest particles are not linear with particle size and would, therefore, not be linear with this plotted ratio. It follows that the ratio of particle size to size of bed interstices is a determinant of filter coefficient only down to particles of a certain "critical" size, and for smaller particles, other factors are dominant.

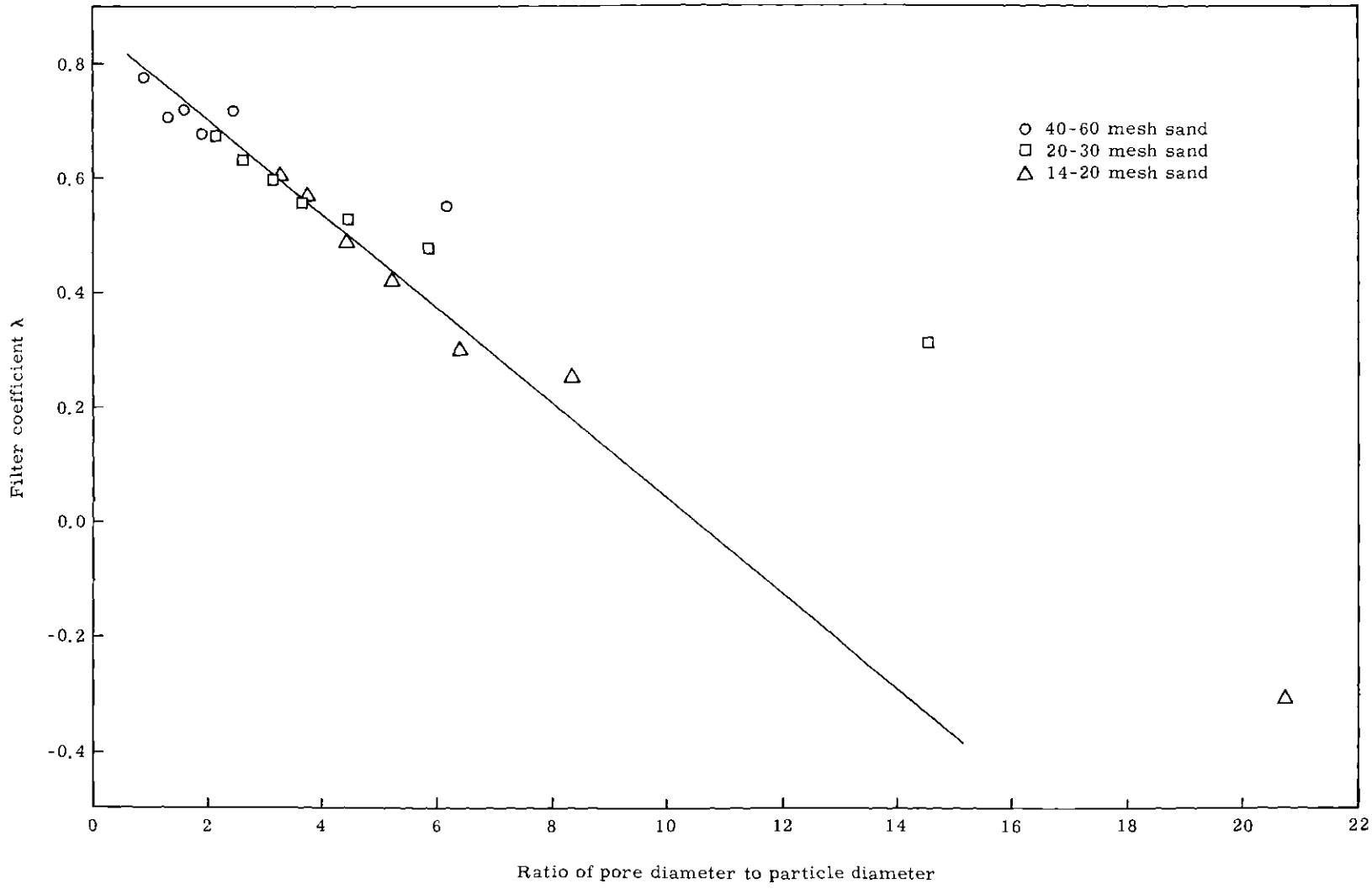

Figure 46. Correlation of Filter Coefficient and Ratio of Suspended Particle Diameter to Maximum Diameter of Sand Constrictions

For the sands evaluated, the transition diameter for particles between domination of relative sizes and other factors is in the range of 25-30 μ. For Anthrafilt, the transition point is around 15 μ.

According to the theory of gravitational sedimentation, the filter coefficient is proportional to the square of the diameter of the particle

$$\lambda = KD^2/\overline{v} \qquad (2.7)$$

Conformance of experimental results to the expectation derived from this hypothesis can be ascertained by plotting λ versus D^2 for conditions of constant velocity. The actual results for λ less than the constant (maximum) value are displayed in Figure 47. It is seen here that a straight line relationship is not present with either Anthrafilt or 14-20 mesh sand. The sparse data for 40-60 mesh sand impair the reliability of the curve prepared from them, but even so there is no suggestion of any proportionality.

The curve for 20-30 mesh sand, when plotted for a square law in D however, does reveal a linear segment which extends over a considerable portion of its length. Taken alone, this curve might lend credence to the gravitational hypothesis, at least for some particle sizes, but when the other curves are included in the evaluation, this is not the case. With the sieving mechanism it is understandable that the particle collection efficiency may vary with particle size, as the probability of removal is dependent on size relationships as discussed above. However, the acceleration due to gravity is constant for all sizes of particles, and if it were the only operative force, all particles should behave in

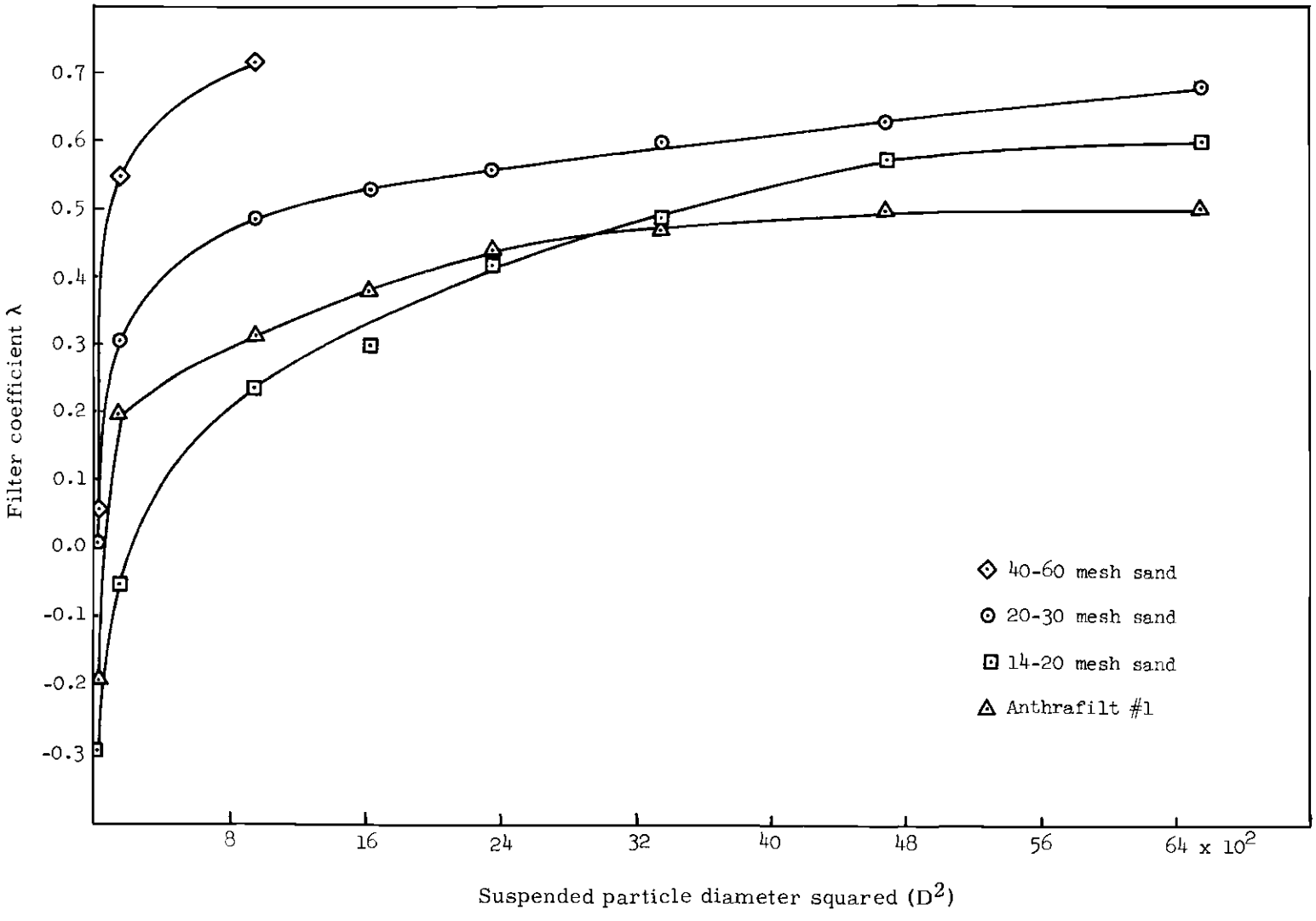

Figure 47. Correlation of Filter Coefficient and Square of Diameter of Suspended Particle at Constant Velocity

similar fashion. A change in the grain size of the bed changes the size of the interstices but does not change the effect of gravity. It therefore appears that gravitational sedimentation at most accounts for only a small portion of rapid sand filtration action. If this force is assigned the symbol f_2 in equation 2.1, its coefficient, B, must be small.

We now consider the situation that exists when a small particle entrained in water is moving near a sand grain surface. If the particle is not of the same density as the surrounding fluid, its velocity will not be identical with the fluid velocity. This difference can be calculated by use of Stokes' law (equation 2.5), but for density differences common to rapid sand filtration the effect would be quite small and, for present purposes, negligible. The simplifying assumption is therefore made that the particle and fluid velocities are identical.

The moving particle is acted upon by several different forces. The gravitational force is acting in the downward direction and it is opposed by the viscous forces operating in the upward direction. The Coulomb force due to double layer interaction provides an attraction or repulsion, depending on whether the charges are alike or not. Even though the charges of the surface and particle have different signs initially, the charge on the surface will become at least partially neutralized by trapped particles. The end result is that most interaction will be required to take place between mobile and arrested particles which have the same sign. For this reason, the double layer force is, for the most part, repulsive.

Attraction due to the van der Waals force is also present, and along with the Coulomb force acts along an axis perpendicular to the

nearest grain surface. The resultant of all the forces determines the direction in which the particle moves and is strongly dependent on the relative positions of the particle and the grain surface. The magnitude of these forces has been calculated for particles of different size under different conditions.

The van der Waals and double layer forces were found from equations 2.13, 2.18, and 2.19, and the gravitational force is the product of the particle mass and the acceleration due to gravity (980 cm/sec^2). The viscous forces vary with the velocity gradient, which is a function of distance from the surface of the sand grain. It is necessary to first determine the velocity profile, and this can be done in the following manner. In laminar flow, the velocity profile is a parabola defined by

$$v = v_c - Kr^2 \tag{5.1}$$

where $K = v_c/R^2$ and v is the velocity at radius r in a tube of radius R. The centerline (maximum) velocity is v_c and is twice the mean velocity of flow.

Let R be 30 μ and mean velocity be 0.4 cm/sec, typical values for rapid sand filtration through 40 mesh sand. Then

$$v = 0.8 - \frac{0.8}{(3 \times 10^{-3})^2} r^2 = 0.8000 - 8.889 \times 10^4 \, r^2 \tag{5.2}$$

The viscous force is then calculated from the equation defining viscosity

$$\tau = \mu \frac{dv}{dy} \tag{5.3}$$

where τ is the shearing force per area, $\frac{dv}{dy}$ is the velocity gradient, and μ is the constant of proportionality. $y = R - r$ and

$$v = 0.8 - 8.889 \times 10^{4} (R - y)^{2}$$

$$= 0.8 - 8.8889 \times 10^{4} (R^{2} - 2Ry + y^{2})$$

$$\frac{dv}{dy} = - 8.889 \times 10^{4} (- 2R + 2y) = 2 \times 8.889 \times 10^{4} (R - y)$$

$$\text{Force} = \mu \times 8.889 \times 10^{4} (R - y) \times \text{area} \qquad (5.4)$$

$$F_{visc} = 17.778 \mu (30 - y) \pi a^{2}$$

(a = particle radius)

Combining constants, ($\mu = 10^{-2}$)

$$F_{visc} = .5585 (30 - y) a^{2} \qquad (5.5)$$

The necessary data for calculation of van der Waals and double layer forces for a number of different experimental conditions are available in the literature.[62,103] Reported studies have included several media and involved waters containing differing amounts of selected electrolytes. The van der Waals forces were calculated by two different equations, but there was at most only a negligible difference between the two results at separation distances where the van der Waals and double layer forces are of similar magnitude.

Table 12 lists the approximate distance of approach between particle and grain surface at which the van der Waals force exceeds the double layer force. The magnitude of forces at this separation distance

Table 12. Separation Distances for Balance of Opposing Forces

Experiment No.	Medium	Electrolyte	Approximate Distance (cm x 10^{-7}) from Surface for van der Waals Force to Exceed Double Layer Force for Particles of Diameter			
			2μ	5μ	10μ	20μ
1	Ballotini	5 x 10^{-5} N KNO_3	1	1	2	3
2	"	10^{-3} N KNO_3	1	2	3	5
3	"	10^{-3} Mg-SO_4	2	3	5	7
4	Anthracite	10^{-3} N Mg-SO_4	1	2	3	5
5	Sand	Tap water	2	4	6	9
6	"	Demineralized Water	6	10	10	20
7	Sand	25 ppm Cl^-	8	10	10	20
8	"	25 ppm $SO_4^=$	6	9	10	10
9	"	25 ppm PO_4^{-3}	3	5	7	10

is shown in Table 13.

The figures of Table 12 show that the critical distance is sensitive to different factors. The major variable is the Coulomb force, which is subject to a possible reversal of direction and is affected by the concentration and valence of the ions present in the water. Thus it may be seen from Table 12 that a five micron diameter particle in water containing 25 ppm Cl^- (experiment #7) will be attracted toward a sand surface when the separation distance is less than 10×10^{-7} cm. A similar particle in water containing 25 ppm PO_4^{-3} (experiment #9) will not be attracted until it approaches to half this distance, 5×10^{-7} cm. Referring to these same conditions in Table 13, it is found that the opposing forces in experiment #7 have a magnitude of 1.04×10^{-7} dyne at the separation distance where they become equal. In experiment #9, the forces are about 8.50×10^{-7} dyne.

From these calculations it appears that any particle closer to a grain surface than a certain critical distance would be attracted and, barring some geometrical peculiarity, would probably be captured. It does not seem impossible, however, that a particle closer to a surface than the critical distance could escape, due to the direction of movement of the water. It is also to be recognized that the direction of particle movement may not be parallel to the surface; it may possess an angular component resulting from the angle at which it traverses the critical point. The geometry may also produce approach to a given surface from above, a situation that would increase the probability of capture as the gravitational force would then act to assist rather than oppose capture.

Table 13. Magnitude of Forces at Separation Distances where Opposing Forces Balance

Particle Diameter (μ)	Weight* (g)	Gram Force (dynes)	Forces at Distance of Balance (dynes)								
			1	2	3	4	5	6	7	8	9
2	1.1×10^{-11}	1.07×10^{-8}	1.67×10^{-6}	1.67^{-6}	4.17^{-7}	1.67^{-6}	4.17^{-7}	4.63^{-8}	2.6^{-10}	4.63^{-8}	1.85^{-7}
5	6.5×10^{-11}	6.37×10^{-8}	1.04^{-5}	2.60^{-6}	1.16^{-6}	2.60^{-6}	6.51^{-7}	1.04^{-7}	1.04^{-7}	1.29^{-7}	4.17^{-7}
10	5.2×10^{-10}	5.13×10^{-7}	1.04^{-5}	4.63^{-6}	1.67^{-6}	4.63^{-6}	1.16^{-6}	4.17^{-7}	4.17^{-7}	4.17^{-7}	8.50^{-7}
20	4.2×10^{-9}	4.11×10^{-6}	1.85^{-5}	6.67^{-6}	3.40^{-6}	6.67^{-6}	2.06^{-6}	4.17^{-7}	4.17^{-7}	1.67^{-6}	1.67^{-6}

*Based on a specific gravity of 1.000.

Whatever the circumstances, if a particle is on a trajectory that causes it to escape the influence of one sand grain, the direction may be such that the particle will be captured on a successive grain. There is also the possibility of deflection away from a surface in passing from the vicinity of one sand grain to the next, but the probability of trajectory change is a function of the exact geometry of the situation and the tortuosity of the flow path and is not amenable to an analytical solution. It seems likely, however, that the probability of deflection either toward or away from a surface is equal.

Based on equation 5.2, the velocity of flow parallel to a flat surface at a separation typical of the critical distance would be about 3×10^{-5} cm/sec, which is a very low rate. A particle moving this slowly would be subjected to the attractive force for a relatively long time, and the probability of escape of a particle from within the critical distance is consequently quite small. It may therefore be concluded that most particles approaching closer than the critical distance would be trapped.

The shape of the curves below the linear region in Figures 28 through 31 can now be explained in terms of these theoretical considerations. According to the theory of double layer interactions, force between suspended particle and grain surface varies directly as the radius of the suspended particle, as shown in equation 2.13. The van der Waals force varies as the square of the radius as shown by equation 2.19. This being the case, in the region of predominance of surface effects, it should be possible to express the filter coefficient in terms of radius and radius squared

$$\lambda = a_0 - a_1 r + a_2 r^2 \qquad (5.6)$$

where a_0, a_1, and a_2 are experimentally derived constants. The forces involved are most frequently working in opposition to each other, and a difference in the sign of the two terms is appropriate. This equation does not include specifically the relationship of bed characteristics, flow velocity, temperature, or other variables and is, therefore, limited to situations where these factors are held constant.

Equations of this type were fitted to the lower portion of each of the curves of Figures 28 through 31, and numerical values for the coefficients were found and are listed in Table 14. Examining the values for the three different sand sizes, it will be noted that both a_1 and a_2 decrease with sand grain size. There is no smooth progression of values for a_0. For Anthrafilt No. 1, the a_1 and a_2 values fall above the largest sand, but a_0 comes between values for 14-20 and 20-30 mesh sand.

Table 14. Coefficients for Equation 5.6

Medium	a_0	a_1	a_2
14-20 Sand	-0.37	-0.062	-0.0015
20-30 Sand	-0.068	-0.081	-0.0029
40-60 Sand	-0.072	-0.14	-0.0056
Anthrafilt	-0.24	-0.045	-0.0047

The a_0 values seem to have the physical significance of representing filter action for particles approaching zero radius. The limit, however, is at the molecular level, and r can never approach zero. It might be assumed that a_0 then represents the capacity for adsorption of solutes, but this would probably be a function of surface area. Relative surface area of grains per volume of bed is not in the same ratio as the calculated a_0 values. Pending the availability of experimental evidence concerning other sand sizes and different media, the a_0 will be considered an experimental constant.

It might be argued that the coefficients a_1 and a_2 represent the relative contributions of the double layer and van der Waals forces, respectively, to the total value of λ, but this does not seem to be the case. The a_2 value, representing the van der Waals portion, is much smaller than the corresponding a_1 value; yet it is usually the unassisted van der Waals force that serves finally to capture and retain the particle. Also, if these coefficients did indicate the division of forces, their relative magnitudes should be the same in each different porous medium, but this was not found to be the case.

It would be useful if the values of the coefficients could be consolidated into a single general equation indicating the relationship of the a_1 and a_2 values to the dimensions of the individual bed grains. It was not possible to devise a meaningful expression for this purpose, and it is therefore necessary to determine values for these coefficients experimentally.

The suspended particles captured by grain surfaces are then seen to be those that come within a certain critical distance of the surface,

but the transport of particles into this zone must also be explained. Two possible mechanisms are worthy of consideration: diffusion and hydrodynamic forces. Diffusion due to random molecular bombardment is a force that is always present and could probably be effective if the particle was small enough and the flow rate slow enough for an extended residence time in the filter. Neither of these circumstances apply, as the smallest typical particle is perhaps two microns in diameter and is several times too large for the Brownian motion to be effective. The flow rate is also much too high.[74,75]

The forces involved in the movement of the water are those which are primarily responsible for the transport phenomenon. In a porous medium, the flow streamlines follow a very tortuous path. Streamlines are compressed at constrictions and diverge in zones of larger cross section. An ideal fluid may be specified as having no rotational component of movement, but real fluids are not irrotational, and this leads to possible displacement of streamlines from their theoretically most probable position. Consequently, a given streamline may easily pass close to a surface at one point and at other locations be far away from grain surfaces.

In the zone of constriction, there is also an increase in the velocity of any matter following the streamline. It is the alternately converging and diverging streamlines that present opportunities for transfer of a particle from one streamline to another and the possibility of reaching a streamline that subsequently passes into an attraction zone where capture becomes possible. Should two suspended particles come together and remain together, the effect would simply be the same

as that for a larger particle. Considering the concentration of particulate matter in water being filtered, this chance contact between particles is considered to be negligible.

The behavior of dissolved activity was examined briefly to obtain a more complete understanding of the action of a rapid sand filter. A dilute solution of cesium-137 was used as described in Chapter IV under the "Effects of Dissolved Activity." As may be seen in Figure 27 there is a rapid build up of activity as the cesium enters and is retained by the filter.

When the concentration of dissolved matter in the water begins to decrease, the equilibrium between dissolved and sorbed matter is changed, and the sand begins to release sorbed material to compensate for the lowered concentration. In this experiment, the capacity of the sand was not exceeded, and the activity level of the effluent reached a maximum of only about one half that of the sand bed. The descending portion of both curves is exponential in form and results from washout of the radioisotope which is sorbed on the sand or held in the almost stagnant water which is trapped in the crevices between sand grains where velocity of flow approaches zero. The sudden onset of decreasing activity in the sand is due to the sudden lapse of activity in the influent.

The net consequences of passage of the dissolved activity through the filter are that the maximum momentary intensity reached by the effluent is much less than that reached within the sand bed, so that the peak effect is spread over a longer period of time. It will be noted that the activity does not return to background level in the period covered

by this figure. Experience during the course of this investigation has shown that this is the usual result. The activity level of a contaminated bed will gradually decrease over a period of many days, but a return to the original background level of unused sand has not been observed following introduction of dissolved activity or uncalcined labeled particles. On the other hand, the use of calcined particles with their extremely high tenacity for the radioisotope leads to little if any persistent increase in background level of the sand.

The passage of dissolved radioactivity through a sand bed produces only transient appreciable effects on the sand, but with Anthrafilt the effect is more lasting. Anthrafilt is an excellent sorption material, and ions that become attached to Anthrafilt are very difficult to remove. This is shown very clearly by experiments involving labeled, but uncalcined, particles. In this condition, the particles lose radioactive ions to the water, producing a situation of both dissolved and attached radioactive atoms.

From the experiment described in "Effects of Dissolved Activity" in Chapter IV, it was found to be nearly impossible to remove cesium-137 activity from Anthrafilt to any appreciable degree. It is possible, however, to remove I-131 although it requires much washing as shown by the figures in Table 4. Additionally, this experiment well illustrates the difference in affinity between Anthrafilt and different radioisotopes. In the practical case of a water treatment plant with an Anthrafilt filter, this introduces another variable with which to reckon: the identity of the radioactive species must be known. If only easily removed isotopes are involved, there is less danger of per-

manent contamination of the filter than with those more tenaciously held by the Anthrafilt. With a persistent isotope, it would probably be necessary to replace the Anthrafilt bed in order to reduce the activity level around the filter to near the original background level.

The significance of the work reported here lies in the experimental evidence presented which helps evaluate certain theoretical concepts. Certainly one must recognize that no theory can be accepted because a limited amount of data is in agreement with expectations based on theory but, conversely, if the data do not fit, serious doubts will be cast on the theory.

The generally accepted exponential law for particle deposition has been shown to be true for narrow ranges of particle size. In the hypothetical case where two different ranges of size are present, it seems likely that each range would follow its own pattern, and the result would be the sum of two exponential functions. Following this line of reasoning leads to the conclusion that a different exponential expression exists for each particle size, and the overall measurable result is the sum of all the individual components. This may account for the exponentially distributed pattern commonly found with particulates that have a very wide range of sizes such as those encountered in commercial operations.

An extension of the present work to cover this instance could be the labeling of different size particles with different isotopes so that the deposition pattern of each could be discerned individually. It is presently not clear whether there would be interference between the different fractions or if each would behave independently.

Here it has been shown that the filter coefficients, as measured, fulfill the predictions of the interstitial sieving theory for particles with diameters of about 20 percent or more of the diameter of the minimum calculated pore constriction. For particles in this size range, there is a linear relationship between filter coefficient and particle diameter. This relationship begins with particles just small enough to enter the pores of the filter matrix and continues down to particles with a diameter in the range of 15 μ to 30 μ, depending on the matrix employed.

As consideration is given to particles of smaller size, there is a transition zone which represents a combination of sieving and other effects. Below this transition size, particles are governed primarily by surface forces. The magnitude of the van der Waals and double layer forces has been calculated in order to determine the separation distance at which the van der Waals force is equal to the double layer force. The equations derived from theory offer a theoretical basis for the shape of the filter coefficient-particle size curves for small particles. It is concluded that small particles are governed primarily by the van der Waals and electrical double layer forces.

The experimental results reported here do not fit the theoretical concept of gravitational sedimentation as a major particle collection mechanism. It is to be noted, however, that sedimentation is a phenomenon that is very sensitive to flow velocity and turbulence, and the negative conclusions regarding this mechanism should be confined to hydraulic conditions similar to those actually used in this study.

These agreements (and lack of agreement in one case) are power-

ful arguments concerning the validity of the mechanisms involved, and it is felt that any alternate theories in order to gain acceptance will have to offer a satisfactory explanation for these results.

This work is also of significance with respect to decontamination of water containing radioactive entities. Passage of dissolved activity through a bed has only a limited effect due to the limited capacity of a medium to adsorb or exchange ions. After the capacity is exceeded, the solution will continue to pass through with little or no additional hindrance. The situation is different where particles are involved, as the presence of increasing amounts of deposited matter resists the flow. If wastes are injected into a subterranean porous formation at the rates used here, deposition profiles should be similar. Under these field conditions, however, backwashing or reverse flow would be undesirable, even if in rare cases it might be possible. It is therefore assumed that deposited solids will not be subject to further movement. A medium having a very low filter coefficient would then be most suitable, as deposits would be spread over a large distance, and the formation would be useful for a long time. With a high coefficient, much matter would be trapped near the point of injection, and further movement of water and particles would be hampered. After particles have come to rest, any subsequent movement of activity will have to occur by separation of active ions or molecules from the stationary particle followed by dispersion due to diffusion or gross movement of the fluid.

The use of a rapid sand filter may be effective in decontaminating certain suspensions prior to release into the environment, but the activity concentrated by the filter may present troublesome problems

of disposal. The tremendous reduction in volume of active matter is very valuable where there is either too much activity or too little water to make dilution a practical solution to the problem.

CHAPTER VI

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

Radiotracer experiments have been performed that confirm certain theories concerning the mechanism by which a porous medium removes suspended particles from flowing water. It has been shown that, in unisize beds such as those described here, suspended particles whose diameter is larger than about 20 percent of the diameter of the pore constrictions of the filter matrix are trapped by a mechanical action, interstitial sieving. This action depends on the size and shape of the individual granules comprising the filter bed but is independent of the chemical composition of the bed material.

Suspended particles of size smaller than 20 percent of the pore constrictions are governed mainly by other forces which depend on the nature and the surface area of the bed grains. The major forces have been shown to be van der Waals and electrical double layer forces. There is no sharp dividing line between the two types of action; there is a transition range of sizes in which both mechanisms are operative. Efficiency of particle entrapment is dependent on both the absolute and relative sizes of both the suspended particles and the grains of the porous medium.

Values of parameters calculated from the sedimentation theory of filter action were compared with experimental values. Theoretical re-

lationships were not confirmed, and the sedimentation hypothesis is rejected for the conditions of these experiments.

Dissolved cesium-137 produces only transient effects on quartz sand but is irreversibly sorbed by ground anthracite.

The filter efficiency measurements made here apply only to unisize filter beds and to suspended particles of narrow size range. Neither of these conditions is normally encountered in commercial practice where bed grains cover a range of sizes and, in addition, a vertical non-uniformity in size distribution results from hydraulic classification caused by backwashing. Suspended particles normally occur in a wide distribution of sizes, and information on the distribution is seldom available. For general applicability, therefore, correction factors are needed to take into account these differences between laboratory and full scale operation. It is hoped to explore some of these areas of investigation in subsequent work.

Recommendations

Since filter efficiency is dependent to some extent on rate of flow through a filter, a logical extension of this work would be to determine values at different flow rates. For a given filter, it would be of considerable practical value to predict the maximum flow rate at which effluent of acceptable quality could be obtained.

Additional work is needed to relate the idealized conditions of the laboratory experiment to the actual conditions encountered in the field. To this end, further filter runs utilizing non-uniform beds and particles with wide size distributions are needed.

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*The abbreviations used herein follow the form referred to in Chemical Abstracts: List of Periodicals Abstracted (1961).

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